

A REPORT ON
NOVEL NATURAL GUM BASED HYDROGELS

A DISSERTATION
SUBMITTED IN PARTIAL FULFILLMENT OF THE REQUIREMENTS
FOR THE AWARD OF THE DEGREE
OF
MASTER OF TECHNOLOGY
IN
POLYMER TECHNOLOGY

SUBMITTED BY

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UNDER THE SUPERVISION OF
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I, Divya Balodhi, Roll No. 2K18/PTE/03 student of M.Tech (Polymer Technology), hereby declare that the project Dissertation titled "**Novel Natural Gum Based Hydrogels**" which is submitted by me to the Department of Applied Chemistry, Delhi Technological University, Delhi in partial fulfillment of the requirement for the award of the degree of Master of Technology, is original and not copied from any source without proper citation. This work has not previously formed the basis for the award of any Degree, Diploma Associate ship, Fellowship or other similar title or recognition.

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ACKNOWLEDGEMENT

The success and final outcome of this project required a lot of guidance and assistance from many people and I am extremely fortunate to have got this all along the completion of this project work. I wish to express my gratitude towards my project supervisor, **Prof. Archna Rani & Prof. Rajinder K. Gupta**, Department of Applied Chemistry, Delhi Technological University, who provided me a golden opportunity to work under their able guidance. Their scholastic guidance and sagacious suggestions helped me to complete the project on time. I wish to thank **Dr. S.G Warkar**, Professor and Head of the Department of Applied Chemistry, Delhi Technological University, for her constant motivation and for providing able guidance. I am thankful to and fortunate enough to get constant encouragement, support and guidance from all teaching as well as non-teaching staffs of Department of Applied Chemistry and Polymer Technology, which helped me in successfully completing my project work. Finally, yet importantly, I would like to express my heartfelt thanks to my beloved family and friends who have endured my long working hours and whose motivation kept me going.

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TABLE OF CONTENTS

CANDIDATE'S

DECLARATION

CERTIFICATE

ABSTRACT

ACKNOWLEDGEMENT

LIST OF FIGURES

LIST OF TABLES

ABBREVIATIONS

1. Introduction

2. Review of literature

2.1. Classification of hydrogels based on various aspects

2.2. General properties of hydrogels

2.3 Polymerization Techniques used for hydrogel synthesis

2.3.1 Bulk Polymerization

2.3.2 Solution Polymerization

2.3.3 Suspension Polymerization

2.3.4 Free- Radical Polymerization

2.4 Natural polymers for hydrogel synthesis

2.4.1 Natural gums and their source of origin

2.4.2 Generalized mechanism of natural gum based hydrogels

2.4.3 Natural gums based hydrogels

2.4.4 Advantages of Natural Gum based hydrogels with respect to synthetic polymers

2.4.5 Limitations of Natural gum based hydrogels

2.5 Applications of Hydrogels in Health care

2.5.1 In Drug Delivery

2.5.2 Contact Lenses

2.5.3 Wound Healing

2.5.4 Scaffolds for tissue engineering

2.6 Applications of hydrogels in agriculture

2.7 Applications of hydrogels in the food industry

3. Methodology

3.1. Materials Required

3.2. Synthesis of Natural gum based hydrogels

3.3. Swelling studies

3.4. Characterization

3.4.1 FTIR Analysis

3.4.2 Thermal Analysis

3.4.3 XRD Analysis

3.4.5 Scanning Electron Microscopy Analysis

3.4.6 Solid-State NMR Analysis

4. Results and Discussions

4.1 Mechanism of formation of hydrogels

4.2 Swelling studies

4.3 Characterization

4.3.1 FTIR Spectroscopy

4.3.2 Thermal Analysis

4.3.3 XRD Analysis

4.3.4 Scanning Electron Microscopy Analysis

4.3.5 Solid-State NMR Analysis

5. Conclusion

6. References

LIST OF FIGURES

SR. NO	TITLE	PAGE NO
Figure 1	Classifications of Hydrogels	21
Figure 2	Swelling Mechanism of Hydrogels	22
Figure 3	Bulk Polymerization Technique	23
Figure 4	Solution Polymerization Technique	24
Figure 5	Suspension Polymerization Technique	25
Figure 6	Hydrogel preparation method	28
Figure 7	Gum ghatti	29
Figure 8	Locust Bean Gum	30
Figure 9	Tamarind Gum	31
Figure 10	Karaya Gum	32
Figure 11	Xanthan Gum	33
Figure 12	Psyllium Gum	34
Figure 13	Konjac Gum	35
Figure 14	Gellan Gum	36
Figure 15	Drug Loading Mechanism	38
Figure 16	Hydrogel utility in contact lenses	39
Figure 17	Wound Healing Mechanism	40
Figure 18	Utility in Tissue Engineering	41
Figure 19	Hydrogels in Agriculture	43
Figure 20	Advantages in food sectors	45
Figure 21	Dynamic Swelling studies	49-50
Figure 22	Dried and Swollen Hydrogels	50-51
Figure 23	FTIR Spectra of Gums and Hydrogels	53
Figure 24	TGA Analysis	56-57
Figure 25	XRD Analysis	58
Figure 26	Surface Morphology of gums and their respective Hydrogels	59-60

Figure 27

^{13}C NMR Spectrum

63-64

List Of Tables

SR. NO.	TITLE	PAGE NO.
Table 1	List of abbreviations	11
Table 2	Depicts hydrogels prepared using various crosslinkers and their mechanism	18-19
Table 3	Natural Gums with their source of origin and chemical composition	26-27
Table 4	Depicts application of Natural Gums based Hydrogels in the Agriculture sector.	43-44
Table 5	Formulation of all Four Natural gum-based Hydrogel along with their Swelling Index (SI).	48
Table 6	Represents SI of all 4 hydrogels in various buffers	50
Table 7	FTIR analysis of all hydrogels of respective gum	52
Table 8	Shows Phase transition of hydrogels and their weight loss %	55
Table 9	Represents the NMR peaks for all hydrogels with their chemical shifts along with functional groups.	61-62

ABBREVIATIONS

LBH	Locust Bean Gum Hydrogel
GGH	Gum Ghatti Hydrogel
TGH	Gum Tragacanth Hydrogel
GAH	Gum Acacia Hydrogel
TGA	Thermogravimetric Analysis
FTIR	Fourier-Transformed Infrared spectroscopy
SEM	Scanning Electron Microscopy
XRD	X-Ray Diffraction
AAm	Acrylamide
MBA	<i>N,N'</i> -methylene-bis-acrylamide
KPS	Potassium per sulphate
SA	Sodium Acrylate

Table 1. List of abbreviations used

ABSTRACT

The future of good research lies in the search of innocuous material development, based primarily on natural resources. Keeping in this perspective, four different natural gums namely, Locust bean Gum, Gum Ghatti, Gum Tragacanth, and, Gum Acacia, were used for synthesizing four different novel hydrogels via free radical polymerization method with sodium acrylate, acrylamide. The synthesis was evident with the help of characterization using Thermogravimetric analysis, Fourier-transform infrared spectroscopy, Scanning Electron Microscopy, ¹³C NMR, and swelling studies in buffer solutions of pH 4, pH 7.4, pH 9.2 and, in distilled water. The synthesized hydrogels are potent enough in its potential utility in pharmaceuticals, drug delivery, tissue engineering as scaffolds that generally mimic human skin and in wound healing activity by incorporating the drug into it.

1. Introduction

In recent decades, rising involvement has been dedicated to the development of hydrogels which are tri-dimensional crosslinked polymers possessing the ability to assimilate water-fluids in it and prevents its dissolution in water due to presence of crosslinks present in them [1]. While more practically, they are very well known for its super absorbent nature and maintain their integrity due to crosslinking in them. These polymeric hydrogels show stimuli when their surroundings such as electric field, pressure, temperature, pH, and even including chemical composition are altered [2,3]. They are classified on the various parameters such as physical properties, preparation, charge, source, and cross-linking between them. Based on the preparation method they are homo-polymeric, co-polymers, and interpenetrating in nature. Based on the charges they are classified as cationic, anionic, and non-ionic. Similarly, based on the source they are natural, synthetic, and hybrid. They are also classified on their physical properties, i.e. smart and conventional. Nowadays, most of the commercial hydrogels are being made of synthetic polymers, thus non-biodegradable and considered as contaminant due to their inappropriate disposal causing the startling situation in the entire world. To uphold an eco-friendly environment, synthetic polymers have an immediate need to get replaced from natural ones as problems of plastic pollution, air pollution, global warming, ozone depletion is causing a threat to the environment as well as human health. However, it is imperative to utilize natural substances as raw materials for different applications to curtail environmental problems as with the passage of time dramatic shift has been seen towards novel materials derived from them. Biopolymeric hydrogels have fascinated many sectors due to their biocompatibility and their harmless nature. They have exhibited novel applications in the field of biomedical i.e. in drug delivery, wound healing, and in a tissue scaffold generation [4]. Their major feasibility has also been seen in the agriculture area as they have been explored as soil conditioners and even they act as a mini reservoir for the soil which retains water and helps improving texture and fertility [5,6]. Presently, a lot of natural polymer based hydrogels have been prepared using various biopolymers.

The limelight of the research is on natural gum-based hydrogels, the reason being versatility of bio-polymers, as incredible improvements have been witnessed as it

serves as novel alternative over synthetic polymers. Technically, they have many advantages which makes it more favourable. They are environmentally friendly, green, biodegradable, renewable, and hence are compatible with nature. They are conveniently available and are generally low cost. They are non-toxic and can easily be modified for improved characteristics [7]. They possess a huge amount of structural diversities. The present work elucidates synthesis, swelling studies, and further crosslinked copolymerization of biopolymer is evident by standard physicochemical methods like FTIR, TGA, XRD, SEM and ^{13}C -NMR of hydrogels based on the following gums, i.e. Gum Tragacanth (GT), Gum Ghatti (GG), Gum Acacia (GA) and Locust Bean Gum (LB).

GT is a natural polysaccharide that usually consists of galacturonic acid, xylose, arabinose, galactose, and fucose in its chain. The presence of carboxylic and hydroxyl groups helps in reacting with the monomer and cross-linker [8–10]. It is a very resourceful bio-polymer and contains two fraction mixture in its main chain, i.e. "tragacanthin", which usually dissipate to give a colloidal solution and the insoluble section i.e. "bassorin", which usually has the potentiality to show swelling in an aqueous medium. The amicable chemistry of GT allows it for the amalgamation of superabsorbent hydrogels [11]. Its potential utility is seen in the biomedical sector as reported by Baljit et al. [12] for wound healing with the help of hydrogel dressing in which they have cross-linked Tragacanth gum with cotton fibre.

GG is a natural polysaccharide and has drawn attraction due to its biocompatible, biodegradable, and non-toxic nature. Its main chain constituents are D-Galactose, D-Glucuronic acid, D-xylose, L-arabinose [13]. It is generally water-soluble and belongs to family Combretaceae. Its most frequent names are dhau, dhawa, raam, etc. It shows its occurrence in the mainly arid forest and is generally glossy [14,15]. It consists of many salt constituents like calcium, potassium, magnesium, and sodium salt. GG based hydrogels have shown their use in pharmaceuticals, controlled drug delivery applications as reported where they have been developed as hydrogels by varying the concentration of crosslinker to the monomer by microwave radiation method [16].

GA is referred to as gum Arabic, which is obtained as gum exudate. Its composition is generally of 1, 3-linked β -D-galactopyranosyl units, L-rhamnose, L-Arabinose, and D-glucuronic acid. It is generally water-soluble [17]. They are generally found in the

west of Africa to generally to the Indian peninsula. GA is the oldest natural gum and is generally recognized as safe (GRAS) and has shown its potential application in the food industry and textile industry. It is also known for its gelling agent, stabilizer and also helps in the extension of shelf life of food. The major applications are seen in biosensors, drug delivery, and even in the agriculture area [18]. LB is a versatile biopolymer which has application in the food and pharma industry and even has potential use in tissue engineering, in implants [19]. It consists of 1,4 linked D-mannopyranosyl units in its main chain [20]. It is generally obtained from seeds while the powder form is produced via milling of the endosperm. It is found in the geographical region of Asia, Africa, and even in South America is also referred to as carob bean gum which belongs to the Fabaceae family. The presence of galactose in its chain enhances its functionality and hence is easy to modify. Fig.1 represents the structure of all-natural gums [21].

MOTIVATION FOR RESEARCH

Mother-earth has gifted humans with the beauty of nature. From early ages mankind has used natural materials, but today the scenario has totally changed and synthetic materials have replaced natural one's causing the alarming situation in the entire world due to its non-biodegradable nature and inappropriate disposes. In order to maintain an ecofriendly environment, synthetic polymers has an urgent need to get replaced from natural ones as problems of plastic pollution, air pollution, global warming, ozone depletion is causing a threat to the environment as well as human health. Hence it is imperative to utilize natural substances as raw materials for different applications in order to minimize environmental problems. Here, we are focusing on hydrogels which have been synthesized using natural gums.

OBJECTIVE

- Synthesis and characterization of Locust bean, Gum Ghatti, Gum Tragacanth and Gum Acacia based hydrogels.

SPECIFIC OBJECTIVE

- The specific objectives which are required to achieve the above objective are :
 - Preparation of LBH based hydrogel, GGH hydrogel, TGH hydrogel and GAH hydrogel using SA and AAm as monomers.
 - Characterization of the hydrogels using
 - TGA
 - FTIR
 - SEM
 - XRD
 - Swelling Studies Using Different Buffers and Distilled Water.
 - ¹³C NMR

2. Review of Literature

Hydrogels are referred to as 3-D polymeric networks which have the competence to swell in water by imbibing it in their voids. They consist of a crosslinking agent in them which prevents the dissolution of the hydrogel in water [22]. The crosslinking can be physical and chemical in nature and is distinguished as in physical crosslinking the bonding is usually interim which is due to hydrogen bonding, electrostatic interactions, etc, and in chemical crosslinking there is permanent bonding between the polymeric chains. Contrasting structures are formed because of the crosslinking of hydrogels i.e. Homopolymers, copolymers, block, and graft polymers [23]. The hydrogels are presumed to hold a huge amount of water in them without losing its structural probity [24]. The expediency of hydrogels in different sectors has attracted due to its flexibility, permeability, and its water retention capacity. The hydrogels' ability to swell is usually due to its hydrophilic group usually being hydrated when in contact with water, so because the water is bound, the hydrogel cross-linked network will swell and expose its hydrophobic groups to proceed contact with water can absorb all the it. The mechanism of swelling is also due to osmotic force which acts as a driving force [25]. They have low mechanical properties, so in order to increase their strength, reinforcing agents are added to build a composite hydrogel. The factors that affect the hydrogel are temperature, pH, solvent composition, magnetic field, and ionic strength. Today, hydrogels have attracted many fields, such as biomedical [22,23], agriculture [26], and food [24]. They also show their use in diapers because they are super absorbent [27]. Natural and synthetic polymers are usually used to synthesize hydrogels according to the application. There are various types of crosslinking agents used to prepare hydrogels. The list is listed in Table 1.

Table 2. Depicts hydrogels prepared using various crosslinkers and their mechanism

Hydrogel	Crosslinkers used	Mechanism	References
Guar gum grafted polyacrylamide	Glutaraldehyde Epichlorohydrin	emulsification method	[28] [29]

Gum acacia crosslinked polyacrylamide	<i>N,N'</i> -methylene-bis-acrylamide	Simple copolymerization	[30]
Gum ghatti based hydrogel	<i>N,N'</i> -methylene-bis-acrylamide	Microwave irradiation	[31]
Gum tragacanth hydrogel	Citric acid	Polymerization	[32]
Locust bean gum hydrogel	Glutaraldehyde	emulsification method	[33]
Gum karaya (GK)-grafted poly(acrylamide-co-acrylic acid)	<i>N,N'</i> -methylene-bis-acrylamide	free radical graft co-polymerization technique	[34]
Tamarind based hydrogel	Citric acid	Cross-linking technique	[35]
Xanthan gum-based hydrogels	Citric Acid	Esterification reaction	[36]

2.1 Classification of hydrogels based on various aspects:

1. They can be classified based on the source of natural polymers, synthetic and hybrid (combination of the two) as illustrated in Fig 1.

A. Natural polymers are classified as polysaccharides, such as cellulose, starch, lignin, xanthan gum, gum arabic, gum ghatti, locust bean gum, and so on. Based on proteins such as collagen, gelatin, etc. They tend to show a highly non-toxic natural, biodegradable and are generally considered environmentally friendly [37].

B. Synthetic polymer-based hydrogels are usually prepared using free-radical polymerization or through an addition reaction in a controlled environment. Some of the most commonly used polymers in hydrogels are polyacrylic acid [38], polyvinyl alcohol [39], etc.

2. Based on cross-linking, they are generally divided into two types, namely physical and chemical cross-linked hydrogels.

A. Physically crosslinked hydrogels are usually formed due to secondary bonds,

such as electrostatic interactions, hydrogen bonds, and hydrophobic interactions between network chains. They tend to have very little energy and therefore have the ability to overcome cross-linking to restore their original form [40].

B. Chemically crosslinked hydrogels are usually formed due to permanent crosslinking between polymer chains. Compared with physically cross-linked networks, they are very stable and therefore have good mechanical properties [41-43].

3. Based on the response, they tend to respond to pH, temperature, light, pressure, magnetic field, etc. Generally, hydrogels tend to change their shape when exposed to different pH buffers. Shrinking, swelling is due to osmotic pressure maintained inside and outside the hydrogel which tends to change at critical pH [44]. Therefore, hydrogels also tend to cause changes in their contraction or swelling behavior, thereby causing stimulation of temperature. At low temperatures, usually, the bonds between the networks are not affected, so the chain is in an extended form. At higher temperatures, the chain becomes weaker and therefore tends to shrink [45]. They are also photosensitive in nature and change their shape due to irradiation with different wavelengths of light [46]. Pressure can also affect the structural changes of hydrogels [47].

4. In terms of ionic charge, they are classified as cationic, anionic, and amphoteric in nature. Hydrogels usually tend to show stimulus towards the electric field due to which counter ions and immobile ions are produced which attracts them towards electrodes either having negative or positive charge namely cationic and anionic [48].

5. In terms of preparation, they can be classified as homo, block, graft, and co-polymer in nature. Generally, they differ from each other due to their composition as homo-polymer is produced due to the reaction mechanism which single type of monomer for example cellulose. In co-polymeric hydrogel usually two types of monomers are used having at least one hydrophilic group in either of the monomer [37]. Block polymers are prepared in which different types of monomers react and get grouped into repeating blocks of every unit of monomer across the polymeric network.

In graft polymer hydrogel preparation one monomer is grafted on the other which usually requires at least two monomers [49].

6. According to degradation, they are divided into biodegradable and non-biodegradable, which usually depends on their degradation rate. Biodegradable hydrogels are usually prepared using natural sources that are rapidly decomposed by microorganisms and are generally environmentally friendly. On the other hand, non-biodegradable hydrogels are usually gels containing monomers or macromolecules, which are difficult to decompose due to their non-degradable nature [50].

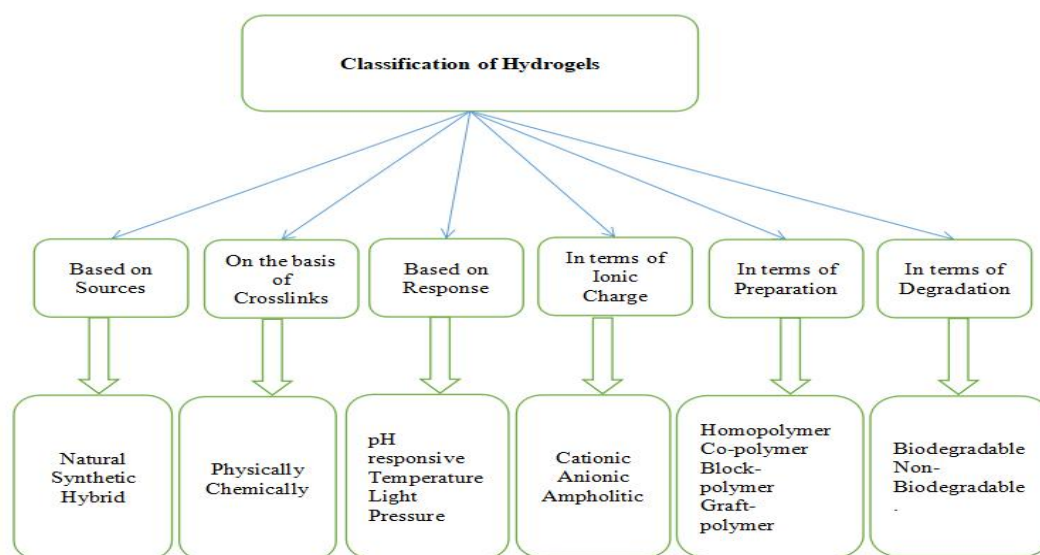


Fig 1. Classification of hydrogels

2.2 General properties of Hydrogels

Hydrogels possess various properties which makes them of its potential purpose that are as follows ;

A) They exhibit great swelling behavior and are elastic due to the presence of more counter-ion groups, which leads to an increase in osmotic pressure and the hydrogel tends to swell [51]. The mechanism is depicted in Fig 2.

B) They have an equal cross-link density distribution in them, causing spatial gel uniformity [52].

C) They show stimulation to pH, temperature, pressure, electric field, magnetic field, and light respectively.

D) They are highly polar and hydrophilic in nature [53].

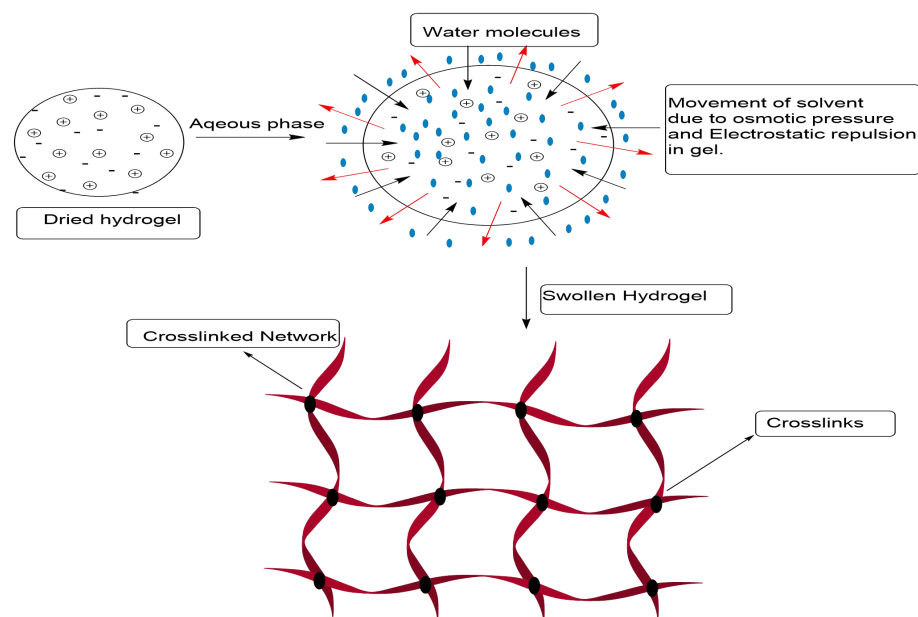


Fig 2. Swelling mechanism of hydrogels

2.3 Polymerization techniques used for hydrogel synthesis:

Generally, natural or synthetic polymers are used to synthesize hydrogels by various techniques, such as bulk polymerization, solution polymerization, suspension polymerization, and radiation polymerization. The explanation of each technique is as follows:

2.3.1 Bulk Polymerization

Bulk polymerization usually requires one or more types of monomer along with a monomer soluble initiator. This technique is quite useful as the high rate of polymerization is achieved in this process due to the presence of a large amount of monomer in it. Usually, the initiator is initiated by UV or by heat treatment. A small amount of crosslinking agent is added for hydrogel formation as shown in Fig 3. The viscosity attained in this process is increased due to the high conversion rate of monomer into polymer allowing more active sites for polymerization with the help of initiator [54]. Generally, hydrogels prepared using this technique possess weak mechanical properties. It is utilized for most advanced development polymers and

numerous kinds of chain-development polymers. On account of chain-development responses, which are commonly exothermic, the warmth advanced may make the response become excessively enthusiastic and hard to control except if productive cooling is utilized. Bulk polymerization has a few preferences over different strategies, these favorable circumstances are the framework is basic and requires thermal protection. It offers various advantages that is the polymer obtained is unadulterated, Huge castings might be arranged legitimately, Atomic weight distribution can be effectively changed, and the product obtained has high optical clearness [55].

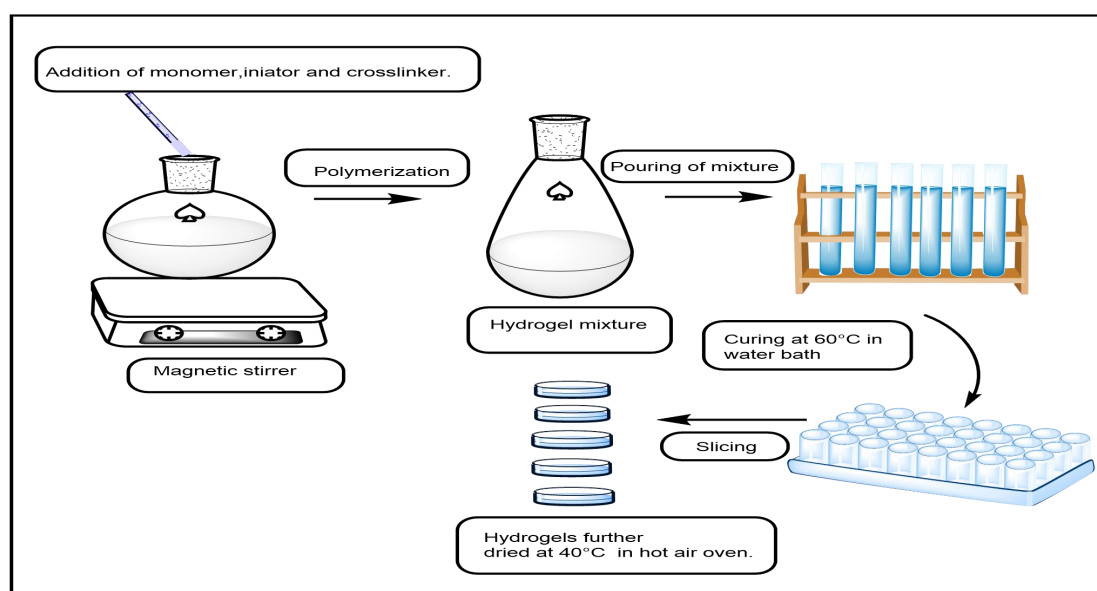


Fig 3. Bulk polymerization Technique

2.3.2 Solution Polymerization

In this method, usually uncharged ions or molecules are selected and allowed to react with an appropriate amount of cross-linking agent. The polymerization is usually carried out by an initiation reaction, in which the initiator is usually allowed to start initiation by means of redox reaction or by UV treatment as shown in Fig 4. Compared to bulk polymerization, this technique is more advantageous because the solvent is a key factor in heat exchange. After preparing the hydrogel, unreacted monomers are usually washed with distilled water [54]. It offers no chain transfer problems.

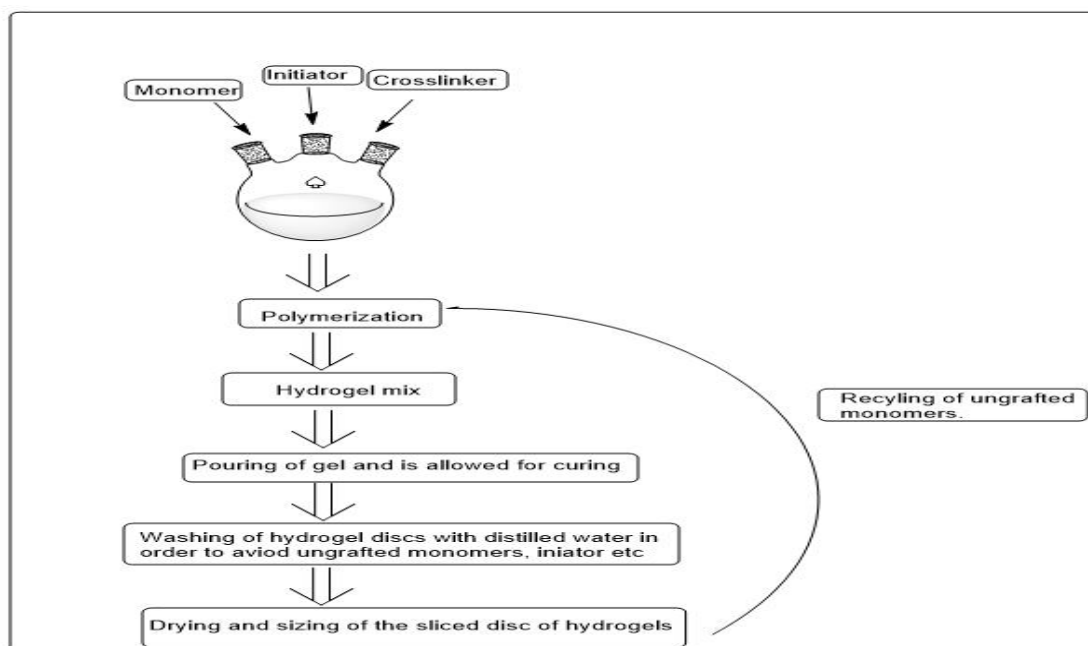


Fig 4. Solution Polymerization Technique

2.3.3 Suspension Polymerization

Suspension polymerization is one of the fruitful techniques used to plan circular or small scale particles with a size scope ranging from 1 μm to 1 mm as shown in Fig 5. This step follows the dispersion method. The monomer and initiator are dispersed in the hydrocarbon phase and allowed to stir at high speed, and further react with the stabilizer in the reactor and form microbeads. Each microbead acts as an independent batch of polymerization. After further adding a cross-linking agent to the medium, they were allowed to prepare spherical hydrogel beads, which facilitated the synthesis of hydrogels. The newly framed small scale particles are then washed to evacuate unreacted monomers, cross-connecting operator, and initiator. The state of particles created can be influenced by the consistency of the monomer stage, while the size of particles can be constrained by the hydrophilic-lipophilic parity (HLB) of each kind of suspending operator [55].

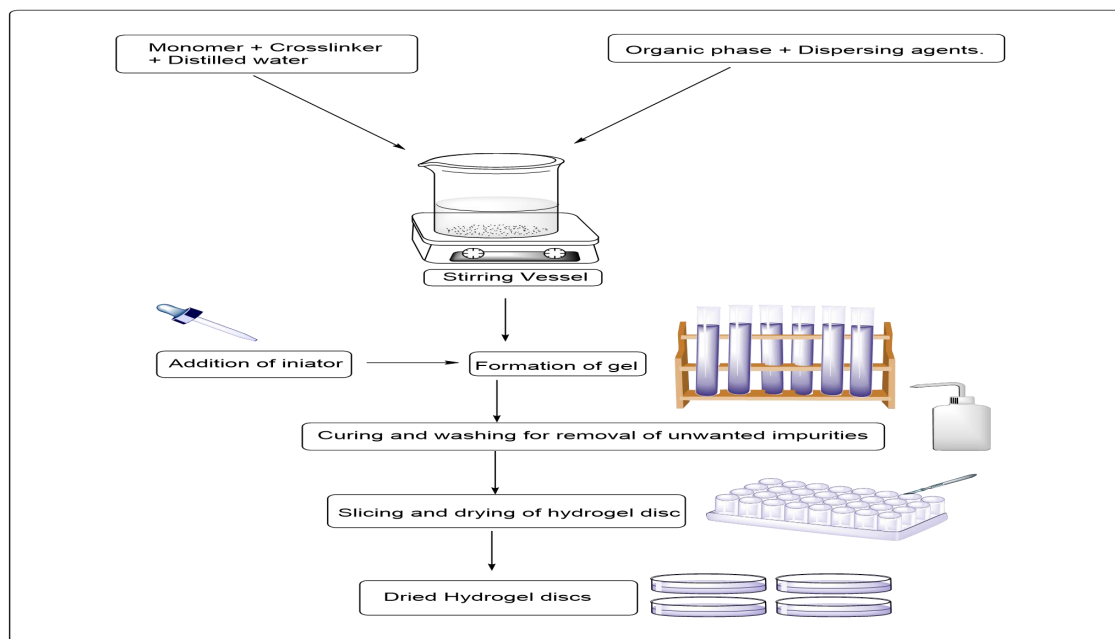


Fig 5. Suspension Polymerization Technique

2.3.4 Free-radical Polymerization:

This technique is superior to other methods because it helps to obtain higher reactivity and can be completed under mild conditions. Initiators are usually used, which decompose with the help of thermal, chemical initiators to form free radicals, thereby attacking the active site of the monomer. The C=C bond is usually included in the main chain. In this type of homopolymer, the copolymer can be formed by changing the monomer type. Free radical polymerization is usually initiated by initiating free radicals, then extending the main chain, and finally terminating. Additionally, radiolysis of water atoms brings about the development of hydroxyl radicals, which likewise assault the polymer chains bringing about the arrangement of large scale radicals. The crosslinking agent helps the hydrogel dissolve by making it a three-dimensional crosslinking network. Examples of hydrogels formed are cellulose hydrogels, dextran hydrogels, etc [56-57].

2.4 Natural polymers for hydrogel synthesis

Bio-polymers are generally versatile in nature. They have origin usually from

microorganisms, plants, and animals. The source of energy which is carbohydrate is usually provided by flora and fauna which gives structural stability to them. Generally, the classification of natural polymers is divided into 6 parts that are protein, polysaccharide, polynucleotides, polyisoprenes, polyesters, and lignin. By comparative analysis generally, natural polymers from synthetic polymer are far better as they possess non-toxicity, biodegradability, and even environment-friendly nature [58-59]. Here in our review the main attraction is towards polysaccharides based hydrogels generally obtained from plant sources which are natural gums.

2.4.1 Natural Gums and their source of origin:

Natural gums have come out to be the most versatile polymer whose source of origin are plants. They have property to easily dissolve in water. They are known to dominate synthetic polymers. They are Eco- friendly, ecological, biodegradable, inexhaustible, and hence are adaptable with mother earth. They are readily available and are generally of modest cost. They are pure and are reliable with nature and it is easy to reorganize its structural integrity. In today's scenario, synthetic polymers have immediate urgency to get retrieved from natural ones as the destruction of the environment is happening which is causing lots of threat to the environment as well as humankind. Hence it is important to utilize the utility of natural gums as basic materials for multiple applications to diminish problems linked to the environment. Currently, natural gums based hydrogels are gaining attention due to its biodegradable nature, compatibly, high water absorption, and painless nature which are very encouraging in various sectors that are in food, agriculture and biomedical area, for example: Guar gum, Gum Tragacanth, Gum Acacia, Locust Bean Gum, Gum Ghatti, Xanthan gum, Tara gum, Tamarind seed, etc. Natural gums are generally referred to as Bio-based polymers. In this review, the main attention is sought by natural gums. Their origin and chemical composition are depicted in Table 2.

Table 3: Natural Gums with their source of origin and chemical composition

<u>Natural Gums</u>	<u>Source of origin</u>	<u>Chemical composition</u>	<u>References</u>

<u>Gum acacia</u>	<i>Acacia senegal</i> , <i>Acacia mearnsii</i>	<u>D-galactose,D-glucuronic acid,L-rhamnose and L-arabinosemin.</u>	[60] [61]
<u>Gum tragacanth</u>	<i>Astragalus gummifer</i>	arabinose,fucose,galactose, galacturonic acid and xylose	[60] [62]
<u>Gum Ghatti</u>	<i>Anogeissus latifolia</i>	D-mannose,L-arabionose,D-galactose,D-galcuronic acid,and D-xylose	[63]
<u>Guar gum</u>	<i>Cyamopsis tetragonolobus (L.) Taub.</i>	β -d-mannopyranose, α -D-galactopyranose	[64] [65]
<u>Tara gum</u>	<i>Caesalpinia spinosa</i>	β -D-mannopyranose, α -D-galactopyranose	[66] [64]
<u>Tamarind Gum</u>	<i>Tamerindus indica</i>	Glucosyl: xylosyl: galactosyl in the ratio of 3:2:1	[67] [64]
<u>Xanthum gum</u>	<i>Xanthomonas campestris.</i>	β -D-glucose, β -d-mannose, α -d-mannose D-glucuronic acid	[68] [65]
<u>Karaya gum</u>	<i>Sterculia urens</i>	Galactopyranosyl and rhamno-pyranosyl with acetyl groups and uronic acid units.	[69-71]
<u>Cassia gum</u>	<i>Cassia tora</i> , <i>Cassia obtusifolia</i>	1,4-(β -D)-mannopyranosyl,galactopyranosyl and glucopyranosyl	[72] [73].

Locust gum	Bean <i>Ceratonia</i> <i>siliqua</i>	β -D-mannose, α -D- galactose.	[64]
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2.4.2 Generalized mechanism of Natural gum-based Hydrogels

The general mechanism for natural gum-based hydrogel is depicted in Fig.6. Generally, natural gum is used in the synthesis of hydrogels preparation. An appropriate amount of gum is used along with monomer in the aqueous phase. The initiator is added to the mixture and stirred with the help of a magnetic stirrer. The crosslinker is added to the mixture for gelation or for forming a crosslinked-polymeric network. Afterward, the mixture is poured into test tubes and allowed for curing at 60 °C in a water bath. After curing they are washed with distilled water in order to avoid unreacted monomers and then are sliced into thin disc and are oven-dried at 40 °C.

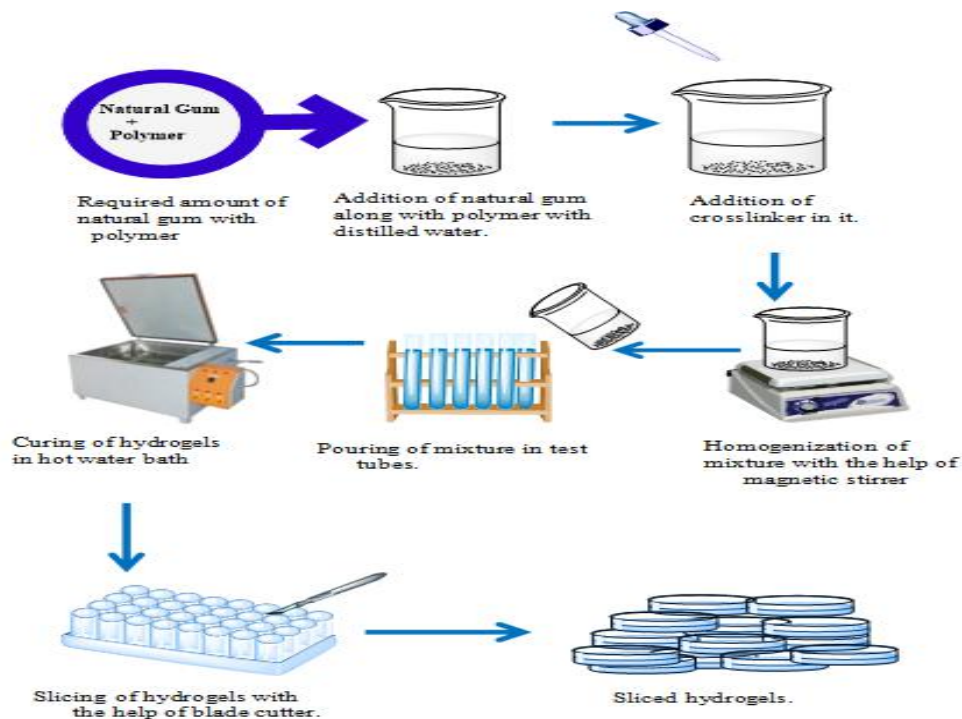


Fig 6. Hydrogel preparation method

Many natural gums are not explored too much extent commercially. The investigation is on the gums used for hydrogel preparation and their applications in various foods, agriculture, health care sector, and their impact on the environment.

2.4.3 Natural gums based hydrogels.

A) Gum Ghatti Based hydrogels:

Gum ghatti is obtained from the plant exudate of *Anogeissus latifolia*. It is an acidic polysaccharide that contains a huge amount of arabinose in its main chain as shown in Fig 7. It comprises of D-mannose, L-arabinose, D-galactose, D-glucuronic acid, and D-xylose in it. It is water-soluble in nature and has white to dark brown colour [63]. These trees are part of the largest forests in India and grow in dry deciduous forests, able to grow under harsh conditions and limited water supply. When provided with nutrients and water, these trees can become very large. When pressure is encountered, Ghatti gum emanates from the tree. In boiling water, it is partially soluble. Gum ghatti based hydrogels has various aspects in food, agriculture, and pharmaceutical industries Table. 3. They were prepared with the help of initiator, and varying concentration of monomers and crosslinkers via Microwave-assisted polymerization reaction as reported by Pinki et al. [31] which involved the dissolution of the required amount of monomer in gum solution with further addition of crosslinker and catalyst resulting in a hydrogel which has application in drug delivery. These hydrogels are generally formed via grafting synthetic monomers or polymers over natural gums. Further, these hydrogels were characterized with the help of TGA, XRD analysis, FTIR, and SEM analysis.

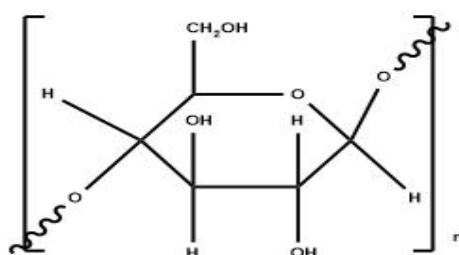


Fig. 7 Gum ghatti

B) Locust bean gum-based Hydrogels:

Locust bean gum is a polysaccharide which is usually derived from plants (Leguminosae) seeds as shown in Fig 8 [74-75]. It is used as a thickening agent, emulsifier, and even in edible coatings in food. The molecular weight of LBG is between 300,000 and 1,200,000 Da. It is almost insoluble in water and requires heating to dissolve it. Because it is nonionic, its water solubility is not affected by the pH or ionic strength of the liquid medium. LBG can be dispersed in hot or cold water to form a sol with a pH of 5.4-7.0, which can be converted into a gel by adding a small amount of crosslinker. The hydrogel preparation by using locust bean gum is reported by Santanu et al [76] using ceric ammonium nitrate as an initiator, in which grafting of acrylamide was done on locust bean via free radical polymerization. The hydrogel was characterized by, differential scanning calorimetry (DSC), Fourier transform infrared spectroscopy (FT-IR) scanning electron microscopy (SEM), X-ray diffraction study (XRD), ¹³C nuclear magnetic resonance (NMR). Hence it is quite evident from the above studies that these hydrogels are biodegradable and non-toxic in nature. Their in-vitro drug release profile was observed to be the same for both of buflomedil Hydrochloride (tablet) and hydroxypropyl methylcellulose. Hence can be used in drug delivery application in health sectors.

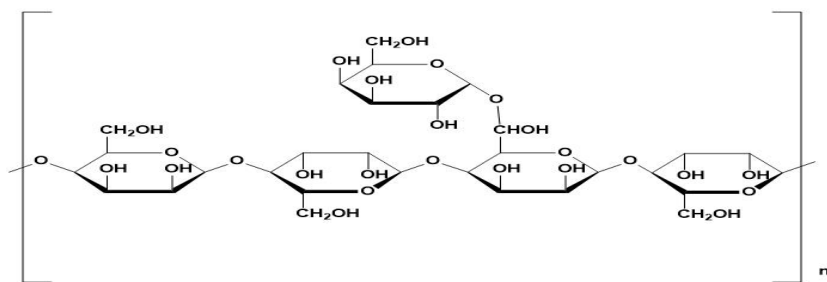


Fig 8. Locust Bean Gum

C) Tamarind gum-based hydrogels:

It is obtained from *Tamarindus indica*. It comprises of Glucosyl: xylosyl: galactosyl in the ratio of 3:2:1 Gerard [67], Sunil et al. [64]. The structural framework of the

tamarind gum skeleton is composed of β -(1/4)-linked glucose units as shown in Fig 9. The sixth position of the glucopyranosyl unit is connected to the side chains of xylose, arabinose, and galactose residues. Enzymatic deportation of the galactose component from the tamarind gum structure also convey thermally reversible gelling ability. The degree of enzymatic cleavage of galactose residues regulate the sol-gel transition temperature. It acts as emulsifying and thickening agents. They are generally acquired by endosperm of seeds. The dissolution is very quick in the water. Its utmost importance is seen in drug delivery by preparing hydrogels with it as reported by Raj et al. [77] in which CNT (Carbon nanotube) were incorporated in various concentration in tamarind gum-based solid hydrogels. The presence of gum was analyzed by FTIR analysis. In-vitro drug study was done using tigecycline as the model drug in which drug release profile was observed with control(having no drug), hence hydrogels with incorporated drug displayed antimicrobial properties. Hence it was summarized from the study that CNT-loaded tamarind gum hydrogel can be explored for drug delivery. They have shown a major contribution towards wound healing and drug delivery application. The main edge of using natural gum is that they are ecologically safe.

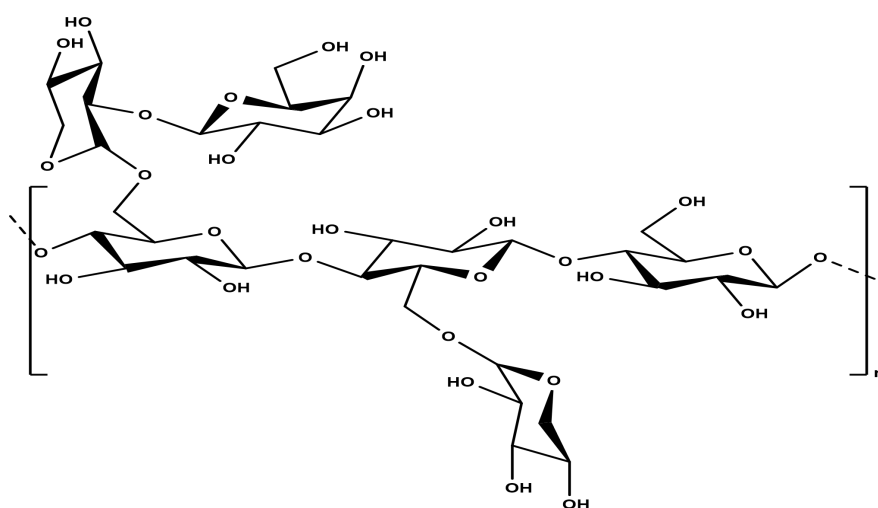


Fig. 9 Tamarind Gum

D) Karaya gum-based hydrogels:

The main chain constituent of gum karaya is composed of galactopyranosyl and rhamno-pyranosyl with acetyl groups and uronic acid units as shown in Fig 10. Usually used in a certain concentration Between 0.2% and 0.4%. In frozen dairy products, it can improve the texture by controlling the formation of ice crystals, thereby further improving its sensory quality. It limits the formation of large-sized ice crystals through aqueous phase management, which results in a smooth texture of frozen dairy products. Its stabilizing effect in low-acid beverages (such as sorbet, fruit ice, etc.) is due to its acid resistance [63]. They are not heat stable. They have shown their major utilization in cosmetics, food industry, and even broader section covers its usefulness in biomedical applications such as in dental adhesives, in epidermal growth of skin which helps in quick healing of a wound. Karaya gum-based hydrogels were prepared by microwave-assisted free radical polymerization technique as reported by Preetha et. al. [78] by incorporation of the apt amount of monomer, initiator and crosslinker with the gum. Polymer-clay hybrid composites were synthesized using modified karaya gum and clay minerals. Before the preparation of clay composites, karaya gum was modified by grafting (2-methacryloyloxyethyl) trimethylammonium chloride. Montmorillonite was used as a clay component. Nanocomposite materials exhibit pH-responsive swelling behavior. The prepared hydrogel composite depicts its efficacious role in eradicating toxic dye removal from contaminated water.

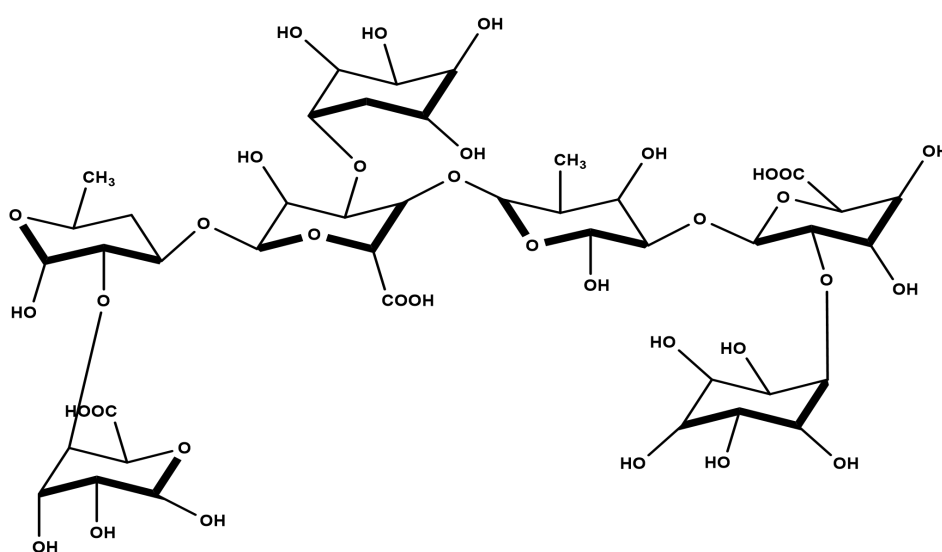


Fig. 10 Karaya Gum

E) Xanthan gum based Hydrogels:

They are prepared by a bacterium *Xanthomonas campestris*. The anionic nature is due to the simultaneous presence of glucuronic acid and pyruvate groups in the side chain as shown in Fig 11. Xanthan gum is durable with a change in external factors such as pH, temperature. As one of them, it has attracted extensive attention. The most successful hydrocolloid, due to its powerful, mainly in key environments such as acidity and high acidity Salt and high shear stress [79]. It is easily soluble in water and hence is used for the preparation of superabsorbent hydrogels which has shown its utility in various sectors including healthcare, agriculture where they have gain importance due to water-retaining properties of hydrogels in soil and even they work as a conditioner for improving texture and fertility of the soil. Xanthan gum is also useful in the synthesis of hydrogels as reported by V.B.Bueno et.al. [80] in which films are prepared via solution casting method with the help of suitable amount of gum and crosslinker in it which are further cured and dried to obtain xerogels. Their role is investigated in drug delivery applications. It has also been reported by Nadia et al. which also explored its utility in drug delivery application by the help of pure polymer chitosan (CS), xanthan gum (XG), monomer 2-acrylamido-2-methylpropane sulfonic acid (AMPS) and initiator potassium persulfate (KPS) through crosslinking agent N’N -Methylene Bisacrylamide (MBA) in the drug delivery of acyclovir (ACV). ACV is a high Effective and selective antiviral drugs for preventing and treating herpes Simple virus (HSV) infection.

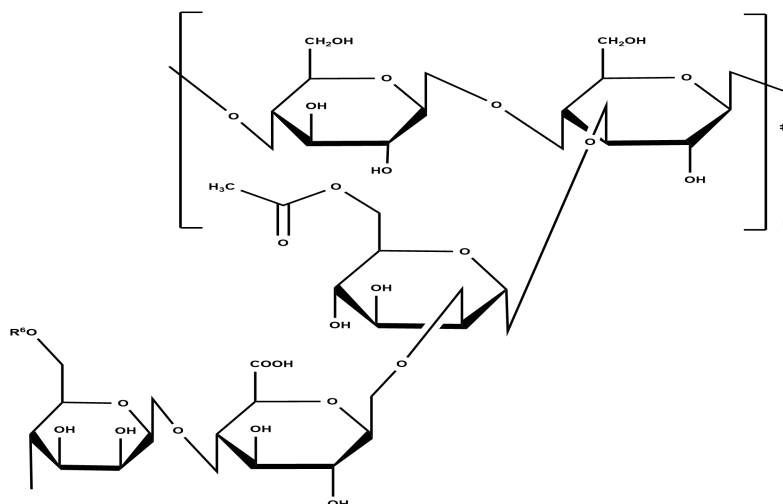


Fig. 11 Xanthan Gum

F) **Psyllium gum-based Hydrogels:**

Psyllium is a familiar name that is used for various members of the plant genus *Plantago*. It mainly comprises of arabinose, xylose, and traces of sugar as shown in Fig 12. They are highly superabsorbent and even environment friendly in nature [81]. Its seeds are mechanically processed by milling/grinding to obtain mucus. It accounts for 25% of the total seed production. Fibrous mucus absorbs about ten times or more of the original water. Transparent, colorless mucus-like gel. Gel properties and composition of polysaccharides extracted from seeds. The Hydrogels were synthesized using gum, initiator, monomer and crosslinker in the reaction flask as reported by J.-K. An et al [82]. The gel obtained is cured and dried at an appropriate temperature. Further, these gels were characterized by FTIR and swelling kinetics was studied [24]. It has shown its promising applications in agriculture and drug delivery sector.

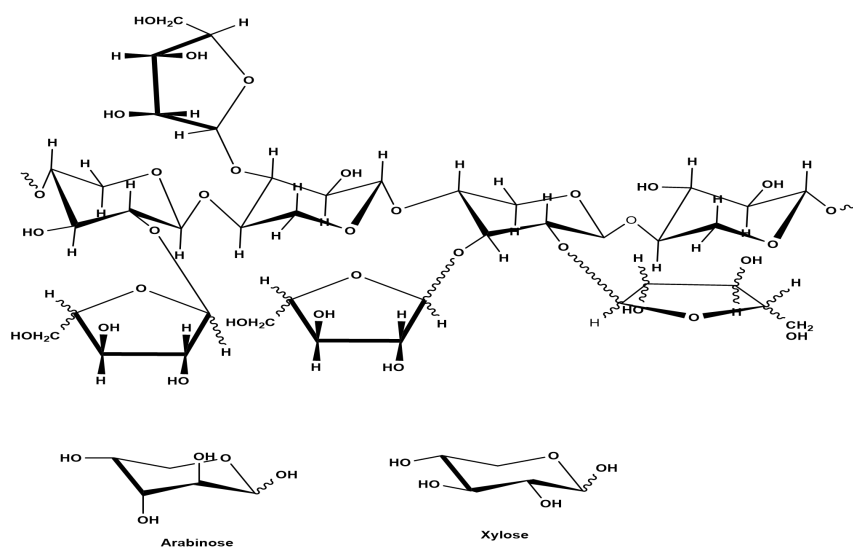


Fig 12. Psyllium gum

G) **Konjac gum based Hydrogels**

Konjac glucomannan backbone comprises of D-Glucose and D-Mannose which is usually linked by β -1, 4 glycosidic linkages as shown in Fig 13 [83]. KGM has been widely used as a gelling agent in the food industry or thickener because of its biocompatibility and biodegradability. The hydrogel prepared from konjac galactomannan is versatile and has attracted towards biomedical applications. The hydrogels using this gum has been reported by L-G Chen et.al in which dissolution of gum along with acrylic acid, catalyst, and crosslinker (N, N-MBA) was added in the reaction flask. After preparation, the resulting hydrogels were washed several times to remove ungrafted monomers from it and further these gels were dried and stored for use. The gels were characterized using FTIR, SEM analysis. Swelling studies, degradation and drug release was also evaluated. The in-vitro release profile of drug 5-ASA was seen and the sensitivity of hydrogels in terms of pH was also observed. Hence it was further confirmed by the studies that konjac glucomannan grafted with acrylic acid can be used as a potential carrier in colon drug delivery [84].

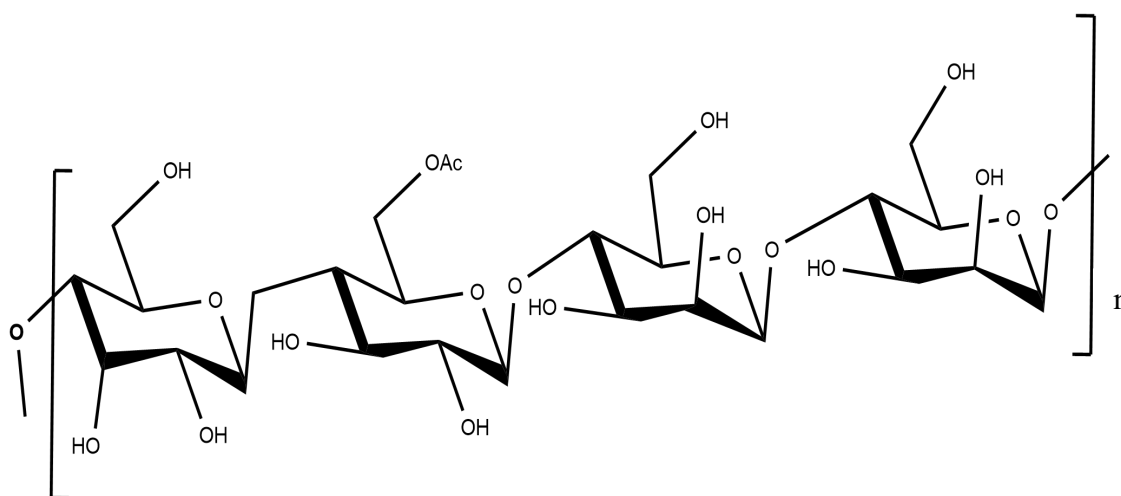


Fig .13 Konjac gum

H) Gellan gum-based Hydrogels

It comprises of glucose, glucouronic, and rhamnose units as shown in Fig 14. It has major applications in the food industry usually as an emulsifier and gelling agent [85-86]. There are two forms of gellan gum which are acetylated (high acyl, HA) and deacetylated (low acyl, LA) which produces thermally reversible hydrogels. Compared with monovalent cations, the gelation of the divalent cation of gellan

gums is more difficult because they promote aggregation through site bonding between pairs of carboxylate groups on adjacent helices. In order to improve its stability or mechanical properties, the functional groups on the gum can be used for chemical modification by photosensitive groups such as methacrylate. Gellan gum-based hydrogels were synthesized using varying amount of monomer and gum in different formulations. Further, these hydrogels were characterized by DMA analysis, degradation, and swelling ability by soaking them in phosphate buffer saline solution up to 170 h. These results validate its potential use in tissue engineering, exhibit non-cytotoxicity as validated from cytotoxicity study conducted on rabbit Nucleus pulposus (NP) cells up to 7 days. MTS assay was done by fusing rabbit NP cells in hydrogel discs. The viability of these hydrogels were also evaluated using Calcein-AM and ATP assay. The study depicted that these hydrogels show potential use in tissue engineering and hence shows the viability of cells. They can even be used as scaffolds as they are reported to mimic the same as the extracellular matrix in humans [87].

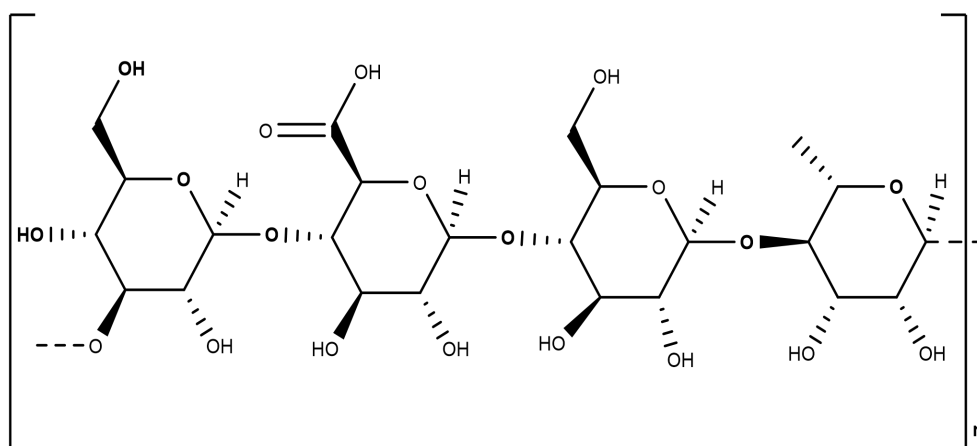


Fig .14 Gellan gum

2.4.4 Advantages of Natural Gum based Hydrogels with respect to Synthetic polymers:

Natural polymers are generally non-toxic and biocompatible as compared to synthetic ones. However the use of natural polymers for pharmaceutical applications are attractive because they are economical, readily available, low cost, non-toxic, and

capable of chemical modifications, potentially biodegradable and also biocompatible in nature [88]. They are a good source for the production of hydrogel-based products due to their renewable nature. On a broader aspect, generally synthetic polymers are generally obtained using petroleum products which demands more cost and shows its limitation due to its non-renewability. Hence to overcome this limitation natural gums are used with synthetic polymers as a hybrid which generally helps in reducing cost and make its availability easier. Generally, they have shown their use in agriculture areas where natural polymers have shown excellent use as they are generally biodegradable in nature and pose less threat to the environment [89].

2.4.5 Limitation of Natural Gum based hydrogels

There are several limitations of natural gum with respect to synthetic ones.

1. They possess weak mechanical properties hence need to get combine with synthetic polymers in order to enhance their functionality.
2. Natural gum usually posses microbial contamination in them due to the presence of moisture content when exposed to the external environment [90].
3. Production of natural gums generally varies due to seasonal and environmental factors [91].
4. The properties also differ due to the growth of gums in various geographic areas and in different climates.
5. Gums when comes in contact with water they possess high viscosity but alteration occurs when they are stored in different atmosphere [92].

2.5 Application of hydrogels in Health care.

There are various applications of hydrogels in health care industries such that drug delivery, wound healing, tissue engineering, biosensors, and even in body implants. Each one is explained below in this review with its potential applications when used with hydrogels.

2.5.1 In Drug delivery

Hydrogels have shown huge applications in drug delivery in the healthcare industry. Drug delivery usually helps to deliver a certain amount of drug loading at a specific

location. The potential use of hydrogels makes drug delivery easier. Generally, hydrogels make it possible by targeted drug delivery at specific sites or controlled release of drugs from low molecular weight to high molecular weight [37]. Hydrogel has attracted medical care because it is easy to trap the drug in the cross-linked network, so it can avoid the influence of external environmental factors, and can easily fix the drug in the gel with the help of external stimuli which are pH, temperature, light and even solvent compositions [93]. Once the drug is incorporated into the cross-linked network, release usually occurs through a variety of mechanisms, including diffusion, release in response to the environment, release through gel swelling, or swelling [94-95]. The drug loading and release curve mechanism is illustrated in Fig 15. The main benefit of using hydrogels in medicine is that it prolongs the release time of the medicine at specific locations where the medicine must be delivered. Natural gum-based hydrogels are generally beneficial because of their non-toxic nature, and are generally more compatible with the human body. Few examples are Guar gum-based hydrogel works as binder, disintegrant, thickening agent, emulsifier, laxative agent, Gum acacia based hydrogel works as suspending agent, emulsifying agent, binder in tablets [60].

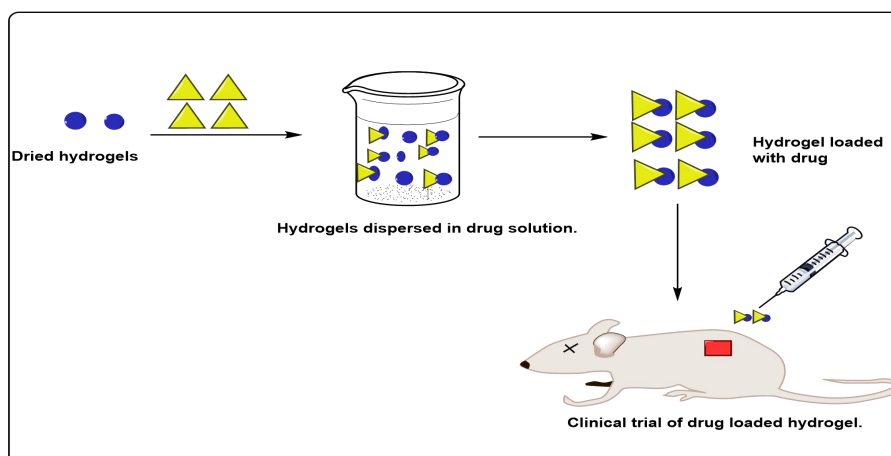


Fig 15. Drug loading and release mechanism

2.5.2 Contact lenses

Hydrogels have also shown its potential utility in contact lenses as displayed in Fig 16. Generally, contact lenses are small devices that are placed on the cornea for alteration in the corneal power. The direct placing of lenses reduced the oxygen exchange which

disturbs the metabolism of an eye which is referred to as hypoxic stress so in order to avoid such circumstances a contact lens must possess greater oxygen permeability through it. Much more mechanical stress occurs in the cornea so to prevent eyes from such stresses the lenses should be made from the right material with proper shape to cover the cornea of an eye [96]. Hydrogels have shown all the properties which make their utility in ophthalmology as they have a property to have a good amount of water content, permeability to oxygen, their non-toxic nature, wettability, and even a good amount of tolerance for the living cells.

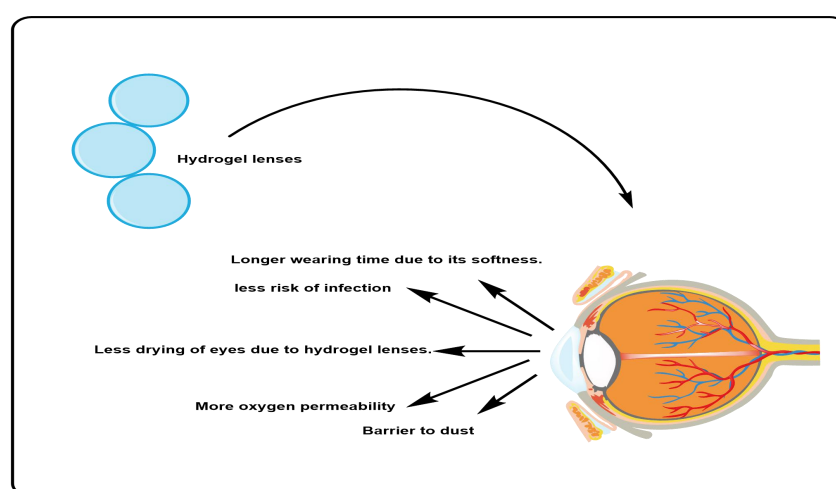


Fig 16. Hydrogels utility in Contact Lenses

2.5.3 Wound Healing

Hydrogels tend to show a diverse and attractive method of removing necrotic and injured cell structures which helps in repairing skin as shown in Fig 17. Generally, dressings are applied to avoid bleeding, protect wounds from infections caused by the environment, by healing it quickly and removing exudates from it. The choice of material is of utmost priority as in case if autologous skin grafts are not available, so in that case, biopolymers may be used as an alternate. Natural gums based hydrogels have gained attraction for wound application due to their availability, non-toxic nature, biodegradability, and its property of mimicking as the cellular environment for the cell and even as a therapeutic reagent which helps in wound healing. Wound dressing synthesized using natural gums has been prepared as reported by Baljit et.al. by incorporating the drug into it which has shown notable potential in sustained drug delivery which supports cell proliferation and therapeutic applications [97]. Therefore,

hydrogel dressings loaded with drugs tend to protect them from infection for a long time and lead to the absorption of wound fluid. Many researchers have reported the use of natural biopolymers in the manufacture of wound dressings i. e. A dressing that uses alginate and gum arabic to make a dressing for further cell proliferation and treatment [98]. Dressings can be used to accurately and continuously release drugs from manufactured dressings [99]. Based on the above evaluations, it can be predicted that natural gums show potential utility in the release of drugs from wound dressings at specific wound sites. Therefore, due to their antibacterial, blood compatibility, and even degradable properties, they can be used in the future. Examples is Psyllium gum-based hydrogels which act in Drug delivery application, curing of diseases, serves as a wound dressing material [72].

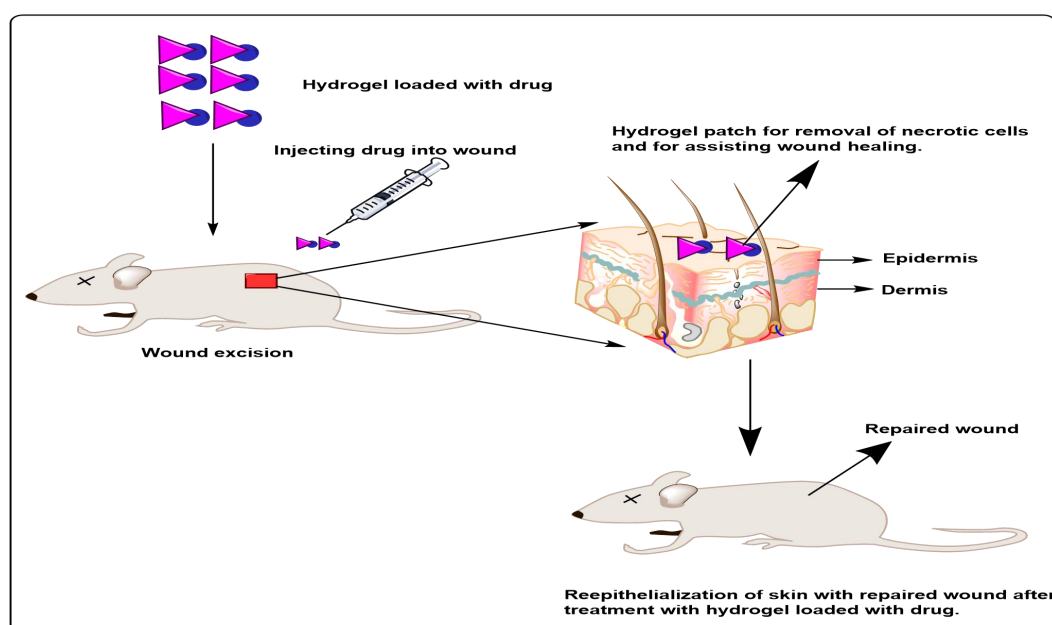


Fig 17. Wound Healing mechanism using drug-loaded hydrogels

2.5.4 Scaffolds for tissue engineering

Tissue engineering is expeditiously emerging as an interdisciplinary area of research aiming to benefit biomedical sectors by providing biomaterials scaffolds that can mimic the cellular environment [100]. Even with the advancement in tissue engineering, yet the response of present medical materials used is not known for a great response as with replacement of the tissue in the human body they tend to show severe cytotoxicity and even hypersensitivity in the cells which lead them to improper functioning inside the human body. Hydrogels as biomaterials have shown its importance in this arena due to their easy modification in mechanical and even

physicochemical properties which are usually somehow like extracellular matrix and have shown their applications in healthcare sectors mainly as biomaterials used in tissue engineering as shown in Fig 18 [101]. Biopolymers have shown their effectiveness as cosmetic agents, and can be used as fillers in scar correction [102], in plastic surgery [81], in neuro tissue engineering and even in bone formation [82]. Hydrogels based on natural gums are biocompatible and biodegradable, which helps to show that the immune system does not have serious reactions, so it has been effective [83]. Few examples are Konjac gum-based hydrogel which act as drug binding material and even serves as a scaffold, Gellan gum-based hydrogel serves as biomaterial in tissue engineering [95, 97].

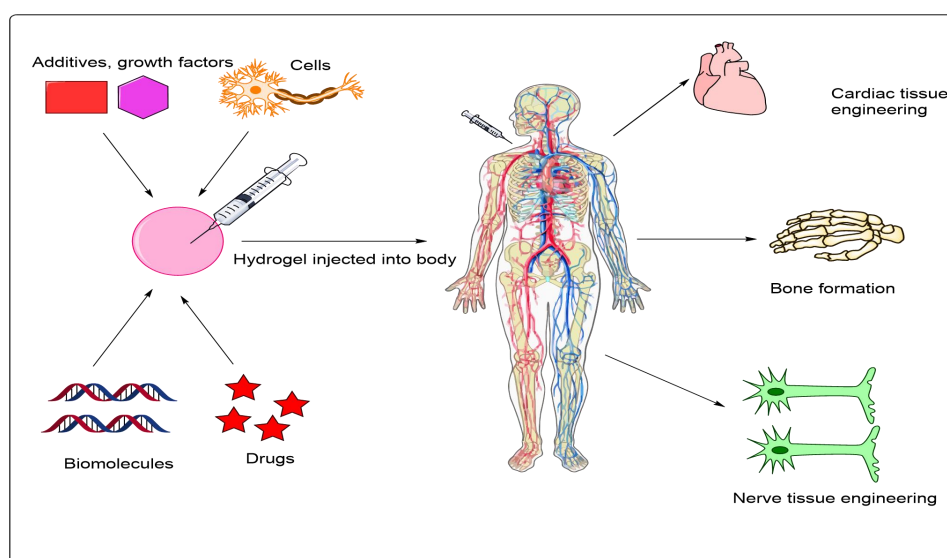


Fig 18. Utility of hydrogels in Tissue engineering

2.6 Application of hydrogels in Agriculture.

In agriculture, hydrogels can be used for micronutrients, controlled release of fertilizers, to increase water consumption and achieve the desired characteristics of agricultural products without harming the environment as shown in Fig 19. The ubiquitous presence of soil enriches the growth of plants and provides them with nutrients. Since the loss of water in farmland has been discovered, which is the main reason for the depletion of crops in farmland, to minimize the loss of agricultural products, hydrogels can play an important role as a reservoir for crops when water is scarce. Porous, it can absorb a lot of water inside [106]. They can help prevent soil

and can reduce irrigation facilities with water crises [107]. Natural gums have shown its utility in agriculture due to their ecofriendly nature as reported by khusboo et.al. where tamarind gum-based hydrogel fabricated with sodium acrylate tends to show more growth as compared with the one without hydrogel tested in chickpea plants. Significant improvement was seen in water retention, moisture absorption and porosity of soil treated with hydrogel as compared to the capacity of untreated soil. Their biodegradability in the soil which was done by soil burial biodegradation test which displays its utmost importance making natural gums the best material choice for hydrogel preparation [108]. Guar gum grafted with acrylic acid with ethylene glycol di methacrylic acid as crosslinker is been reported by Nandkishore et al. Exhibits biodegradable properties and its presence improved porosity, moisture absorption, and water absorption up to 800 ml water per gram. They synthesized hydrogel affirms its utility as a soil conditioner for agriculture use [109]. Hence, hydrogels favor agriculture sector by displaying them as:

1. Superabsorbent crosslinked gel for crop enhancement.
2. Assist in discharge of fertilizers in agricultural fields
3. Helps in increased availability of water for prolonged crop survival.
4. Boost the use of fertilizers and minimizing water contamination.
5. Halting in water runoff hence leading to lower soil erosion
6. Intake of heavy metal ions which helps in less toxicity for crops.
7. Enriched germination for favorable growth of plants.
8. Increased microbial activity.
9. Improved soil moisture and fertility.

In future alluring properties of natural gum-based gels will be admissible in agriculture means with superior gel durability, non-toxic, reasonable price, and for its harmless nature. Diverse applications of natural gum-based hydrogels in agriculture sectors displayed in Table 4.

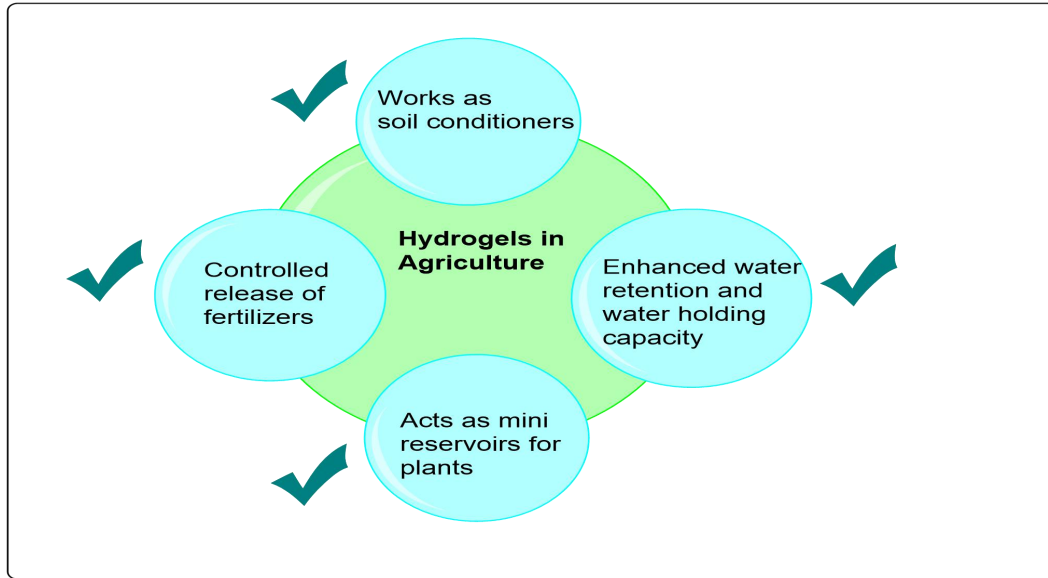


Fig 19. Hydrogels in agriculture sectors

Table 4:Depicts application of Natural Gums based Hydrogels in the Agriculture sector.

Natural Gums Based Hydrogels	Application in Agriculture area.	References
Gum Acacia Grafted Hydrogel	water-retention and moisture preservation.	[110]
Gum Tragacanth Hydrogel	high water absorption And act as mini-reservoir for plants.	[111]
Gum Ghatti Hydrogel	Enhanced water retention, Increased moisture retention	[112]
Xanthan Gum Hydrogel	controlled-release fertilizer, act as water reservoir	[113]
Cashew Gum Hydrogel	Soil conditioning, more water holding capacity	[114]
Tamarind Gum Hydrogel	Maximum water holding	[108]

	capacity, water retention capacity	
Guar Gum Hydrogel	water retention, Soil conditioners	[109]

2.7 Application of hydrogels in the Food industry.

The misuse of petroleum-based polymers i.e. synthetic polymers have created immeasurable damage. Their non-biodegradability has created heaps of waste in the environment due to its improper disposal which has caused an alarming situation all over. So, to minimize its effect, the influx of biopolymers in the food packaging industry is a much-needed function. Biopolymers as they are known for their biodegradability, non-toxicity, and naive nature [115]. Various biomaterials (natural gums, polysaccharides) are used for food packaging but resulted in lower mechanical properties and poor barrier to moisture using guar gum alone as packaging material as reported by Tang et.al [116]. They offer various advantages to food sectors as shown in Fig 20. Many modifications were done but were not apt in a cost point of view. Hence to avoid limitations hydrogels were introduced in this sector which resulted in controlled moisture leading to a reduction in food spoilage [117-118]. In the food industry hydrogels can be utilized in food packaging by building barriers to moisture, microbes hence reduces food spoilage and increases its storability [119]. Natural gums based hydrogels show its utmost importance in the food industry due to its harmless nature as reported by Bandhopadhyay et. al.by using guar gum and bacterial cellulose in polyvinyl pyrrolidone – carboxymethyl cellulose (PVP-CMC) film. The introduction of guar gum tends to improve the mechanical and barrier properties of the film. They displayed outstanding elastic and load-bearing capacity and hence are designed as a substitute used as a biodegradable film due to its degradation study which resulted in an 80% loss of film in 28 days in vermicompost [120].

Bounteous usage of gums based hydrogel is seen in the food sector as modified gum arabic based hydrogel used in hydrolysis of lactose and lactose contained in milk by achieving lactose-free milk by immobilization of β -D-galactosidase without significantly losing its enzymatic activity [121]. Hence, in

future hydrogel based on natural gums can be used in the food industry in various outlets i.e packaging, in edible film, seed coating for improvement in its life span as these gums are generally low-cost and are renewable and hence are harmless to our environment. Minimum literature is available for work on natural gum-based hydrogels in the food industry and hence provides an open window in this arena for more research in the future.

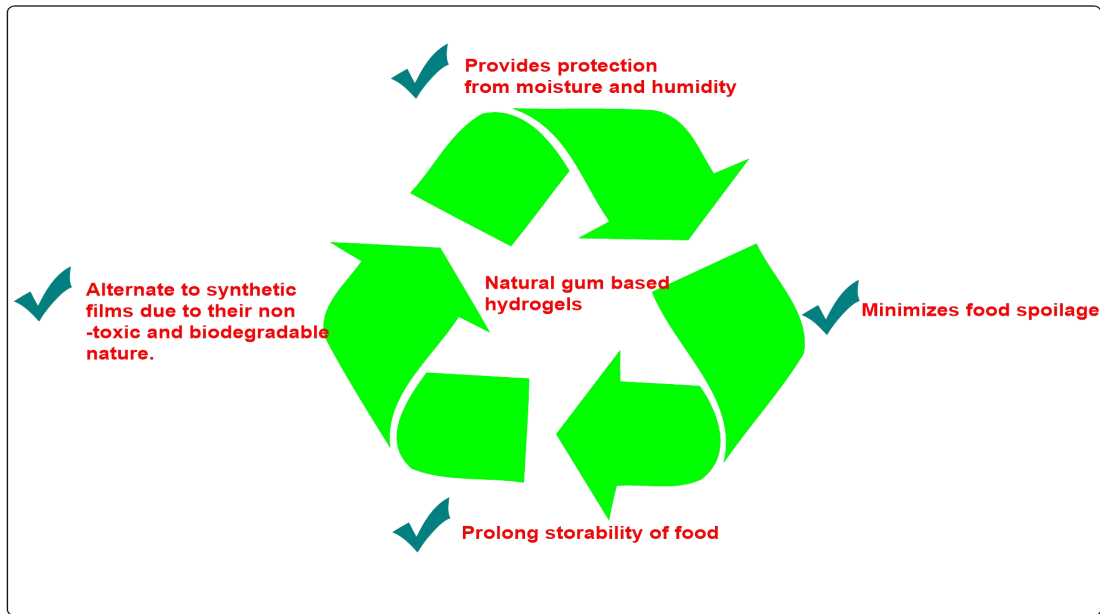


Fig 20. Advantages offered by hydrogels in Food sectors

3.METHODOLOGY

3.1 Materials Required:

Locust bean Gum (LG) was purchased from (Himedia Laboratories Pvt. Ltd, Mumbai, India), (Tragacanth Gum (TG) was purchased from Loba Chemie Pvt. Ltd), Gum Acacia (GA) (Sigma Aldrich, USA), Gum Ghatti (GG) (CDH, New Delhi), Acrylic Acid (AA, CDH New Delhi), Acrylamide (AM, CDH, New Delhi), N, N'-methylene bisacrylamide (N, N-MBA, CDH, New Delhi), Potassium Persulfate (KPS, Fischer Scientific, Mumbai), Sodium Hydroxide (Fischer Scientific, Mumbai) were used. Distilled water (DW) was used as a solvent in the procedure.

3.2 Synthesis of Natural Gum Based Hydrogels

The hydrogels were synthesized using all four natural gums one by one. In brief, all set of hydrogels was prepared by free radical polymerization mechanism. In this methodology, the hydrogels were prepared by dispersing the prefixed amount of natural gum, SA, acrylamide in 50 ml DW separately for all four gums. Afterward KPS and N, N-MBA were added to the beakers to homogenize solution using a magnetic stirrer. After stirring for 2h, the mixture was transferred into test tubes and was allowed to cure at 60°C in a water bath. Then these cured hydrogels were sliced into thin discs and were washed using DW to remove unreacted monomers. Finally, these discs were air-dried at room temperature and further dried at 40°C for 3-4 days till constant weight was achieved in a hot air oven.

3.3 Swelling studies

Swelling studies were conducted using a buffer solution of pH (4, 7.4 and 9.2) and DW at room temperature. The dried hydrogels of each gum were weighed using weighing balance and were immersed in the solution. Periodically, each swollen Hydrogel was taken out and excess of water was wiped off using filter paper. The swelling index (SI) was calculated for each gum, according to the given formula:

$$SI = \frac{W_{SG} - W_{DG}}{W_{SG}}$$

Where, W_{SG} is the weight of swollen gel, and W_{DG} is the initial weight of dried gel. The experiment was conducted for all four hydrogels namely Locust bean hydrogel

(LBH), Tragacanth gum hydrogel (TGH), Gum Ghatti hydrogel (GGH), Gum Acacia hydrogel (GAH) in triplicate.

3.4 Characterization

3.4.1 FTIR analysis

FTIR spectra of natural gums (LB, GG, GT, GA) was done by preparing the KBr pellet (by Perkin Elmer spectrum version 10.5.3). The hydrogels (LBH, GGH, TGH, GAH) were analyzed using ATIR.

3.4.2 Thermal analysis

The TGA (Thermogravimetric analysis) was recorded for each hydrogel with its gum derivative using a Perkin Elmer in N₂ atmosphere. The temperature range was from (25°C to 900°C) with 10°C/min.

3.4.3 XRD Analysis:

XRD analysis was done for each gum and its hydrogel using Bruker D8 ADVANCE, angle ranging from 10° to 80° with an increment of 0.04° and scan speed of 0.5 Sec/step.

3.4.4 Scanning Electron Microscopy

SEM analysis was done for the examination of surface morphology of gums and hydrogels respectively, using JOEL JSM -6610LV.

3.4.5 Solid-state NMR

¹³C NMR was carried out for all four hydrogels and its derivative gums using model JOEL Resonance ECX-400.

4. Results and Discussions

4.1 Mechanism of formation of Hydrogels

Hydrogels were synthesized using KPS as initiator which was decomposed at 60°C to produce free radicals. These sulfate free radicals attacks on the carboxyl groups of natural gums (-COOH) and became active in the reaction. The gums by donating hydroxyl groups (-OH) and formed covalent bonds with sodium acrylate and acrylamide causing propagation and hence polymerizing them into poly (sodium acrylate) and poly (acrylamide). Then cross-linker MBA, (CH₂(CHCONHCH₂NHCOCH) CH₂), was added to the reaction mixture which did the cross-linking between poly (sodium acrylate), poly (acrylamide) and natural gum. This resulted in the formation of the interpenetrating three-dimensional polymeric structure. The prepared hydrogels formulation is given below in Table 5.

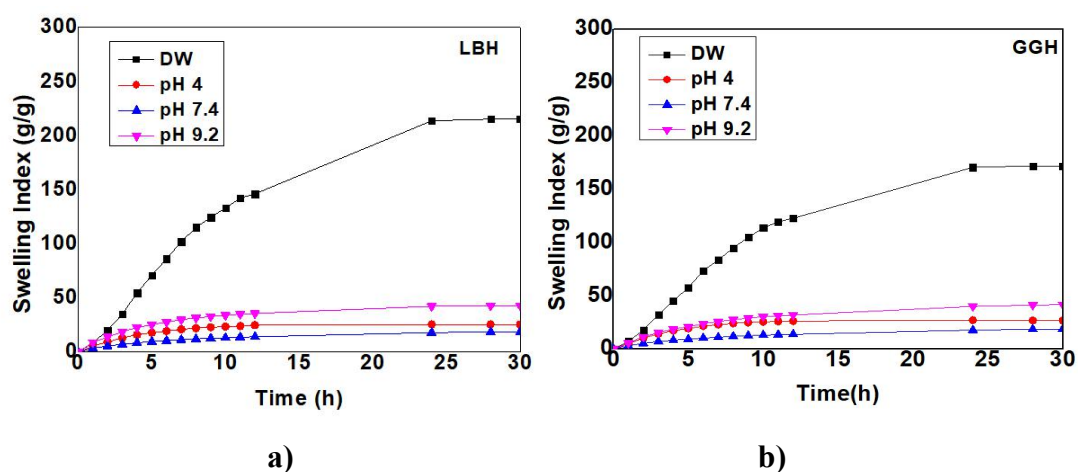
Table 5. Formulation of all Four Natural gum-based Hydrogel along with their Swelling Index (SI).

Gums Used	Gums (gm)	Monomer (AA) mL	NaOH (gm)	Monomer (AM) (gm)	Crosslinker (N, N-MBA) (mg)	Initiator (KPS)/DW (gm/mL)	SI in DW (g/g)
(LB)	0.5 gm	5 mL	3.2gm	5 gm	60	0.05/10	215 g/g
(GG)	0.5 gm	5 mL	3.2gm	5 gm	60	0.05/10	171 g/g
(TG)	0.5 gm	5 mL	3.2gm	5 gm	60	0.05/10	168 g/g
(GA)	0.5 gm	5 mL	3.2gm	5 gm	60	0.05/10	203 g/g

4.2 Swelling studies

The swelling index (g/g) to time (h) is for all 4 hydrogels and is compiled in the form of graphical representation via Fig.21. The swelling studies were conducted in (DW), different pH buffer (4, 7.4, 9.2). The swelling equilibrium for all the hydrogels was observed for about 28 (h). Interpretation of the Swelling index (SI) in distinct solvents was recorded highest in the case of DW for all the hydrogels, which are 215 g/g, 171 g/g, 168 g/g, 203 g/g for LBH, GGH, GTH and GAH respectively. Interestingly, all

four hydrogels showed a similar pattern of solvent absorption where SI tends to increase from (pH 4-pH to 6) and then decrease and further increasing to pH 8 [122]. For LBH, at pH 4, SI was 25 g/g followed by pH 7.4 in which SI was observed to be 18 g/g whereas, at pH 9.2 it was 43 g/g. Similarly, for GGH at pH 4 it was 27 g/g, at 7.4 it was 19 g/g and at pH 9.2 it was observed as 41 g/g. For TGH, the values of SI for pH 4, pH 7.4, pH 9.2 were 24 g/g, 17 g/g and 37 g/g and respectively. For GAH, the SI for pH 4, pH 7.4, pH 9.2 was observed as 27 g/g, 16 g/g and 40 g/g. According to the above evaluation, LBH was observed to have shown maximum SI in DW and hence has more pores for water absorption as compared to others. The increase in the transition of swelling from pH 4 to pH 6 could be attributed to electrostatic repulsion of the hydrogel which results in drastic swelling at this pH range. Whereas, swelling tends to decrease in an alkaline medium which may be due to the presence of excess sodium in the media, which results in shielding carboxylate anions ($-\text{COO}^-$) present in the hydrogels which might prevent anion-anion repulsion between them [123]. Again transition in SI is seen at higher pH, which is at 9.2 where the hydrogels show drastic swelling as compared to pH 7.4 due to conversion of ($-\text{COOH}$) groups into ($-\text{COO}^-$) and causing electrostatic repulsion between them and dissociating hydrogen bonds [108]. The dried and swollen hydrogels of respective natural gums are shown in Fig.22. The tabular form represents SI of all 4 hydrogels in various buffers in Table 6.



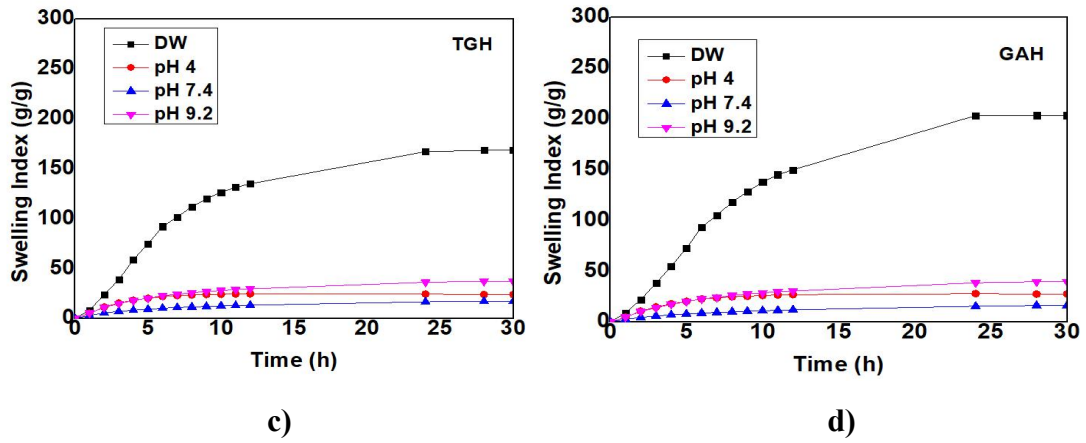
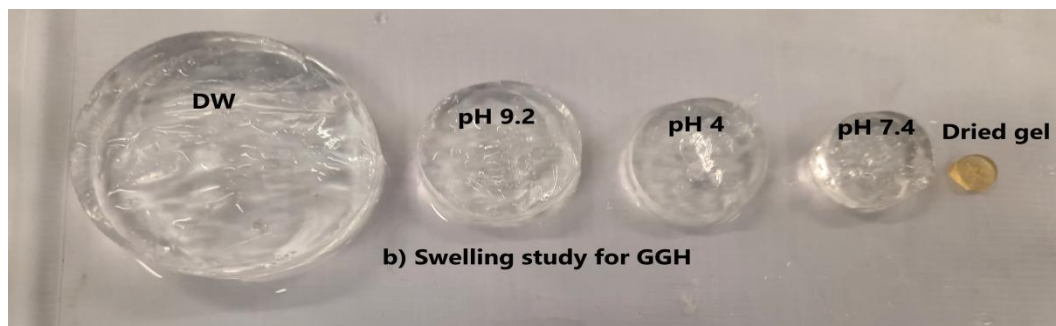
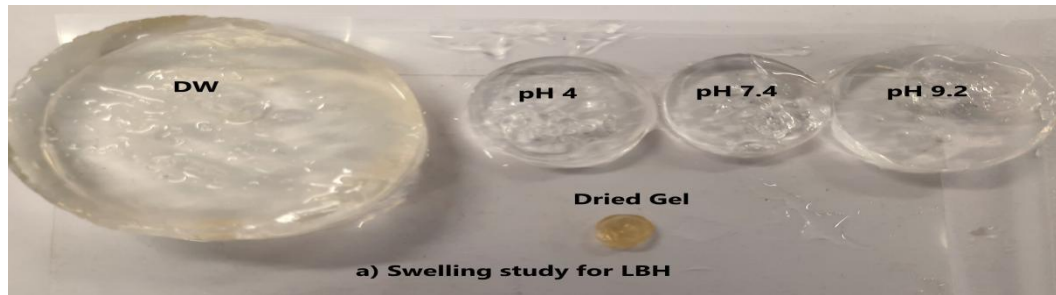


Fig.21. Dynamic Swelling Studies a)LBH b)GGH c)TGH d)GAH

Table 6: Represents SI of all 4 hydrogels in various buffers.

Hydrogels	SI in pH 4 (g/g)	SI in pH 7.4 (g/g)	SI in pH 9.2 (g/g)
LBH	25	18	43
GGH	27	19	41
TGH	24	17	37
GAH	27	16	40



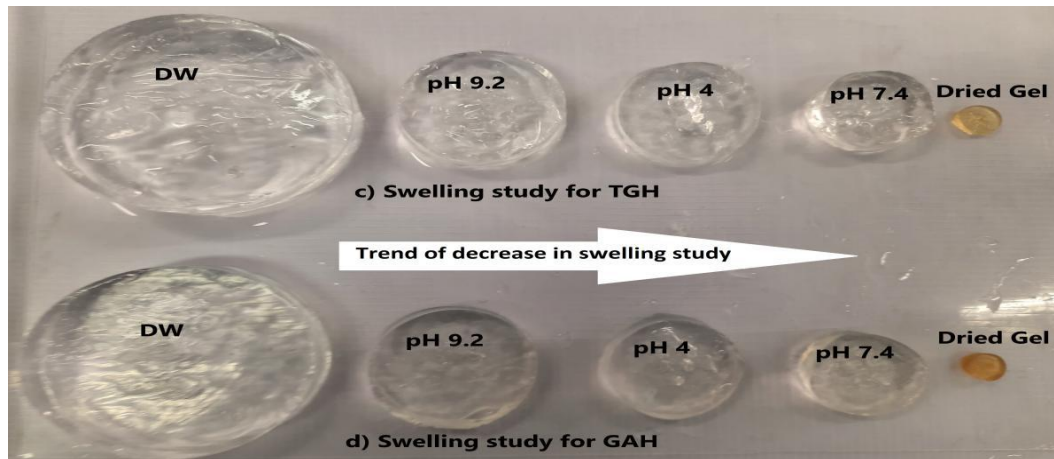


Fig.22. Dried and Swollen Hydrogels in various solvents

4.3 Characterization

4.3.1 FTIR Spectroscopy

The FTIR analysis for all gums and their hydrogels is shown in Fig.23. The peak at 3450 cm^{-1} emphasized (-OH) group for LB, which responsible for hydrogen bonding in the gum. The peaks, i.e. 2900 cm^{-1} (C-H), 1384 cm^{-1} (C-H stretching) and 1145 cm^{-1} (-OH bending) shows symmetrical vibration. For LBH, the peak at 3334 cm^{-1} represented hydroxyl group (-OH) but the reduction in the intensity of frequency depicted crosslinking in the hydrogel as compared to the gum. The peak at 1663 cm^{-1} (C=O), 1033.36 cm^{-1} (C-O-C) showed the presence of the amide linkage due to poly(acrylamide), and it was evident that the reaction between the monomer and crosslinker had taken place [124]. The Peak at 3433 cm^{-1} showed (-OH) in GG. Similarly, peak at 2992 cm^{-1} and 1102 cm^{-1} was due to (C-H) stretching and (C-O) bending. Further peaks at 1551 cm^{-1} and 1626 cm^{-1} showed the presence of (-COOH) symmetric and asymmetric vibration and hence confirmed the IR of GG as reported earlier. Similarly, GGH showed the presence of (-OH) group at peak 3338 cm^{-1} . The strong peak at 1798 cm^{-1} was observed, which attributed to absorption shown due to the linking of poly (sodium acrylate) to gum depicting (C=O) functional group. Further confirmation of hydrogel was evaluated by peak band from ($907\text{-}1340\text{ cm}^{-1}$) which showed (C-N stretching) and confirmed the presence of cross-linking with N, N-MBA, and poly (acrylamide) [16]. In GT, the absorption peak for (-OH) group was also observed at 3423.26 cm^{-1} . The crucial peaks were seen at 1625 cm^{-1} which was due to the carboxyl group while 1743 cm^{-1} was due to the presence of the carbonyl group in the gum as reported earlier. The peak for TGH, i.e. 3423.26 cm^{-1} showed the

presence of (-OH) group. The peak at 1720 cm⁻¹, 1553.25 cm⁻¹ depicted (C=O), and (COO⁻) asymmetric vibration and assured the cross-linking of monomer on natural polysaccharide [125]. GA, also showed (-OH) functional group having a peak at 3398.70 cm⁻¹. The peaks from (1000-1074.6 cm⁻¹) showed (C-O-C) symmetric and asymmetric vibration, thus authenticated the functional group present in the gum that has been reported. GAH also showed hydroxyl group, but with the drop in its intensity as interpreted by comparative analysis with the gum i.e. 3336.10 cm⁻¹. Amide linkage was seen at 1660.99 cm⁻¹ (C=O) and 1555.79 cm⁻¹ (C-O) asymmetric stretching was observed, which justified the presence of poly (sodium acrylate) and poly (acrylamide) in the hydrogel [126]. The tabular form is represented below in Table 7.

Table 7: FTIR analysis of all hydrogels of respective gum

Hydrogels	Wave number (cm⁻¹)	Functional group
LBH	3334 cm ⁻¹ 1663 cm ⁻¹ 1033.36 cm ⁻¹	Due to presence of (-OH) group due to hydrogen bonding in the gum. (C-O) stretch (C-O-C) stretching due to presence of amide linkage in poly (acrylamide).
GGH	3338 cm ⁻¹ 1798 cm ⁻¹ 907-1340 cm ⁻¹	Due to presence of (-OH) group (C=O) stretching due to linking of poly (sodium acrylate) to gum C-N extending
TGH	3423.26 cm ⁻¹ 1720 cm ⁻¹ 1553.25 cm ⁻¹	Presence of (-OH) group (C=O) stretching due to crosslinking with the gum. COO ⁻ asymmetric vibration
GAH	3336.10 cm ⁻¹ 1660.99 cm ⁻¹ 1555.79 cm ⁻¹	Presence of (-OH) group (C=O) stretching due to crosslinking with the gum. (C-O) asymmetric stretching

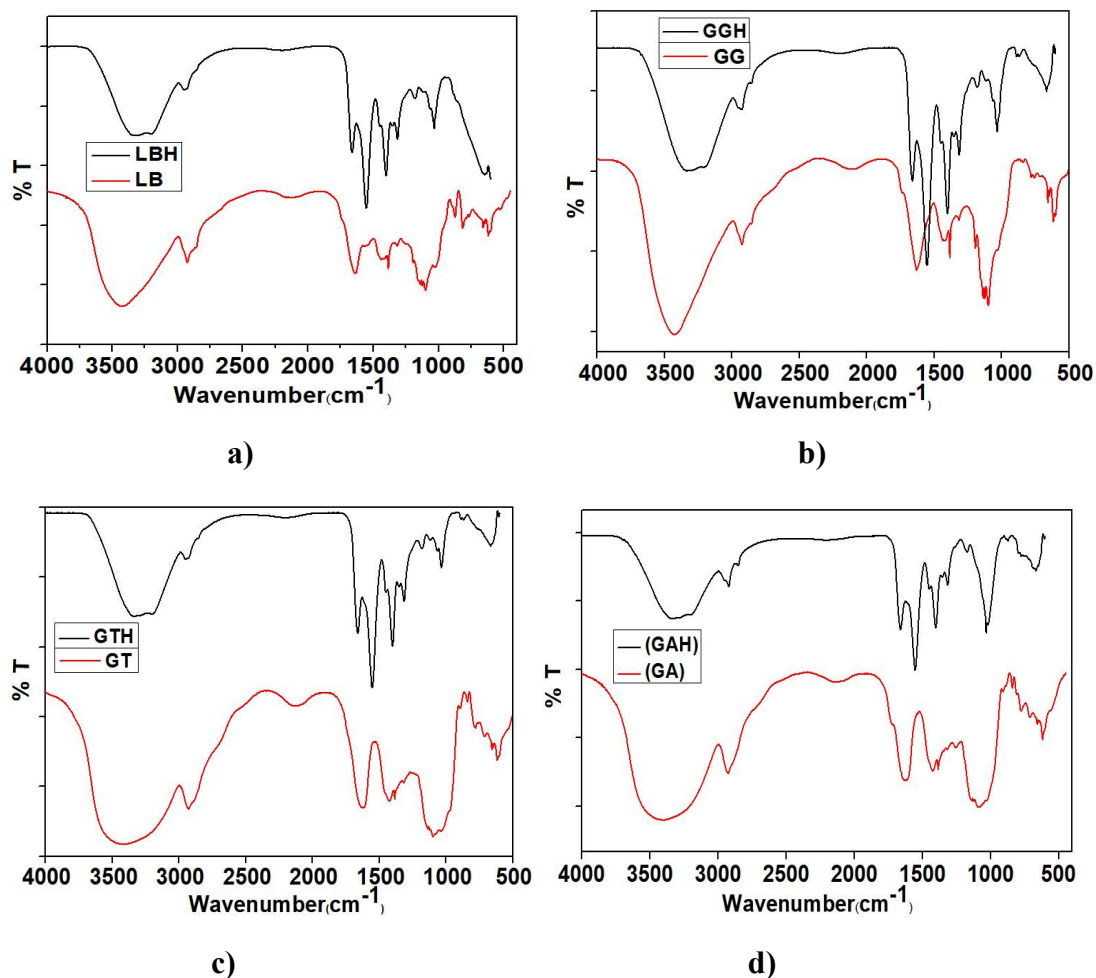


Fig.23. FTIR Spectra of Gums along with their hydrogels a) LB & LBH b) GG & GGH c) GT & GTH d) GA & GAH

4.3.2. Thermal Analysis

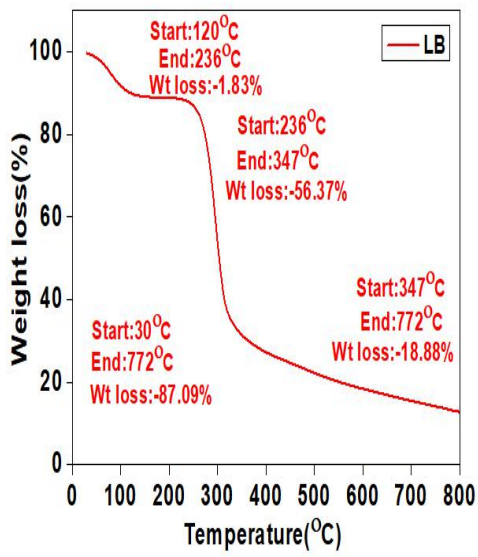
The thermal stability of hydrogels tends to improve as compared to natural gum. So to verify, analogous thermal studies were done. TGA graphs of all hydrogels with their gums are displayed in Fig.24. In LB, weight loss was 1.83% in the initial period (120-236°C) which could be associated due to loss of moisture from the gum. The second phase (236-347°C) of weight loss was 56.37% that might be due to the degradation of the main chain constituents. The third transition was seen to have a weight loss of 18.88 % at (347-772°C) which could be responsible due to further decomposition of glycosidic linkages in it [127]. In LBH, the initial decomposition occurred at (202-446°C) with a weight loss of 38.98% due to entrapped water in it. The second transition (446-513°C) shows 9.76% weight loss & the third phase (513-772°C) weight loss% was off 11.88% because of degradation of polymeric

chains of cross-linked networks. Hence, in comparison of LBH to LB, LBH seems to have more thermal stability as residual mass left at 722°C was 31 % and 13 % respectively from which it is interpreted that modification of gum has led to increasing in its degradation temperature. Similarly, for GG two-phase decomposition was observed first in (223-329°C) and second at (329-772°C) with a weight loss % of 42.64 & 26.51 % due to moisture removal and degradation of biopolymer. In GGH, a three-phase decomposition pattern was observed as for the initial stage (184-425°C), second stage (425-524°C), third stage (524-772°C) that was 33.21, 16.42 and 11.6 weight loss%. The residual mass leftover at the final decomposition temperature for both GG and GGH was 14.14 and 33.01%, which clearly illustrated that thermal stability has been improved for GGH [16]. For TG and TGH three-phase decomposition occurred. For TG, it was for an initial period (223-317°C), the second stage (317-428°C), the third stage (428-772) depicted the weight loss % that was 46.93%, 13.4%, and 23.85%, respectively which could be due to water removal at the initial stage and the degradation of bio-polymer at second and third transition [12]. In TGH, the initial period (190-440°C), the second stage (440-508°C), the third stage (508-772°C) displayed weight loss % that were 38.68%, 11.28% and lastly 13.064%. The residual mass left at the final temperature (722°C) for Gum and hydrogel was approximately 1% and 30% respectively from which it is evident that alteration in the arrangement of polymeric networks has enhanced the thermal stability of the modified gum. In GA, also three-phase decomposition was observed from (119-230°C) with weight loss of 2.74% due to (-OH) hydroxyl group removal. The second transition was from (230-348°C) which portrayed 55.14% weight loss which was due to degradation in the main chain. The third decomposition observed was from (348-772°C) with a 21.35% weight loss. Similarly for GAH, the initial stage was from (183-433°C), the second stage (433-579°C), followed by the third stage (519-772°C) which illustrated weight loss of 37.78 % due to moisture removal, 12.18% in the second phase and 17.1% in the third phase respectively due to disruption in cross-linked polymer network due to degradation with temperature respectively [128]. The residual mass left at the final temperature was recorded more than 19.16 % for GAH than GA which validated more thermal stability of GAH over GA. These observations clearly showed that thermal stability has increased in the case of all

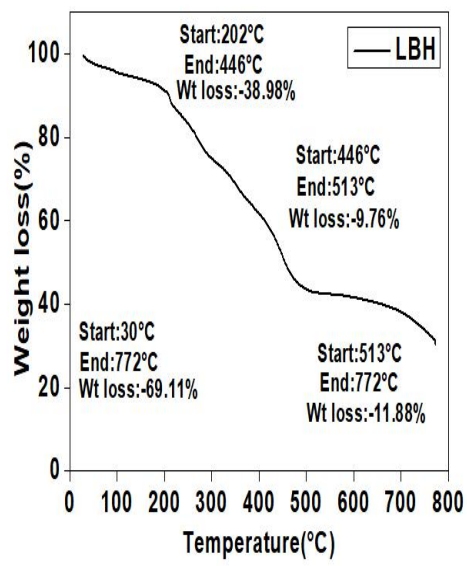
hydrogels as compared with their native natural gums. The tabular form is represented below in Table 8.

Table 8: Shows Phase transition of hydrogels and their weight loss %

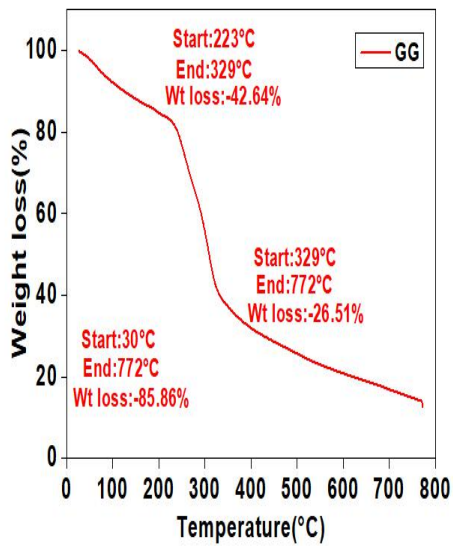
Hydrogels	Time period	Weight loss %	Reason
LBH	202-446°C	38.98	Due to entrapped water
	446-513°C	9.76	Degradation of polymeric chains
	513-772°C	11.88	
GGH	184-425°C	33.21	Due to evaporation of water from the hydrogel.
	425-524°C	16.42	Due to disruption in cross-linked polymer network
	524-772°C	11.6	
TGH	190-440°C	38.68	Due to moisture removal
	440-508°C	11.28	Degradation of polymeric chains
	508-772°C	13.064	
GAH	183-433°C	37.78	Due to moisture removal
	433-579°C	12.18	Due to disruption in cross-linked polymer network
	519-772°C	17.1	



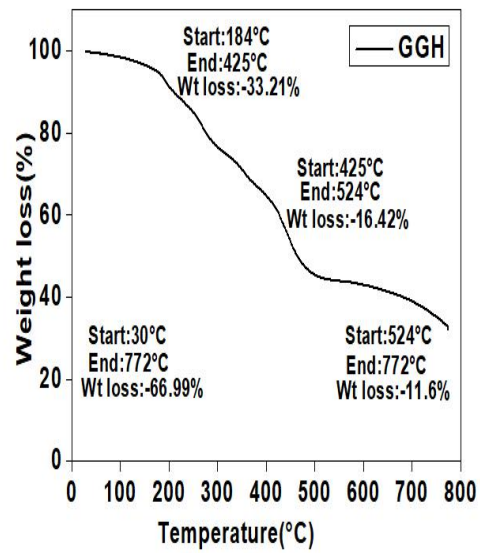
a)



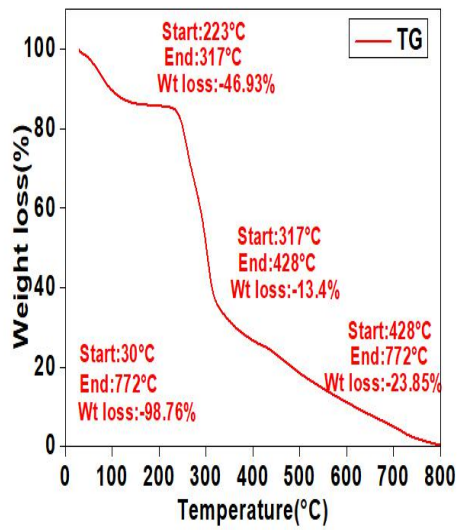
b)



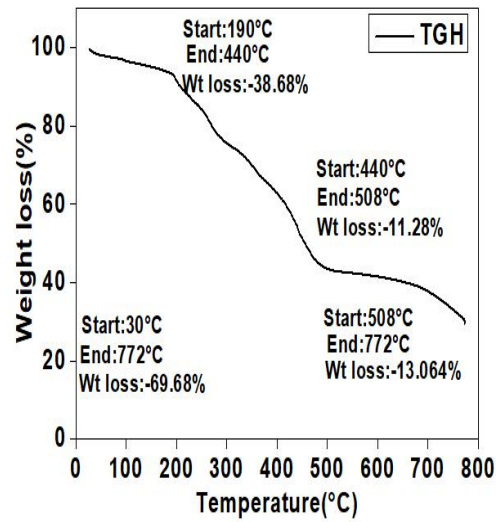
c)



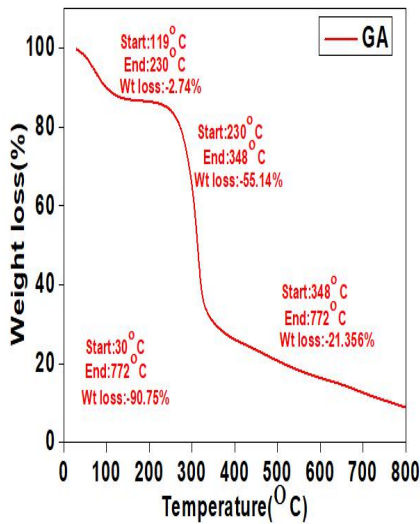
d)



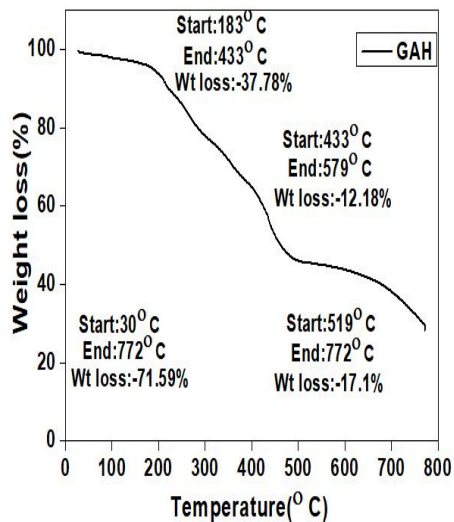
e)



f)



h)



g)

Fig.24. Depicting TGA Analysis of a) LB b) LBH c) GG d) GGH e) GT f) GTH g) GA h) GAH

4.3.3 XRD Analysis

XRD is usually done for the analysis of crystalline, semi-crystalline, and amorphous nature of the material. The XRD plot for all gums with their hydrogels is portrayed below in Fig.25. Indigenous gums have shown a more sharp peak compared with their respective hydrogels. In LB, the arrangement depicted a sharp peak at 19.08°, which showed its semi-crystalline nature. In LBH, a broadened peak was examined at 23.88°

which depicted its amorphous nature which was due to transformation in LB due to crosslinking in it [124]. The plot of GG showed a strong peak at 19.18° which indicated its crystallinity as reported earlier [16]. The deviation in the peak of GGH was seen after crosslinking that affirmed its amorphous nature which might be due to disorientation in the structural arrangement of the crosslinked hydrogel at 24.11° [30]. In TGH, 21.72° was observed that showed modification in peak due to the formation of new networks, which reduced the interaction among TG chains which resulted in declining of semi-crystalline nature when compared to its native gum [12]. In GA, 19.36° was interpreted which represented semi-crystalline nature, but GAH showed variation in peak (27.88°) due to the modification of the GA in GAH due to co-polymerization and crosslinking [130].

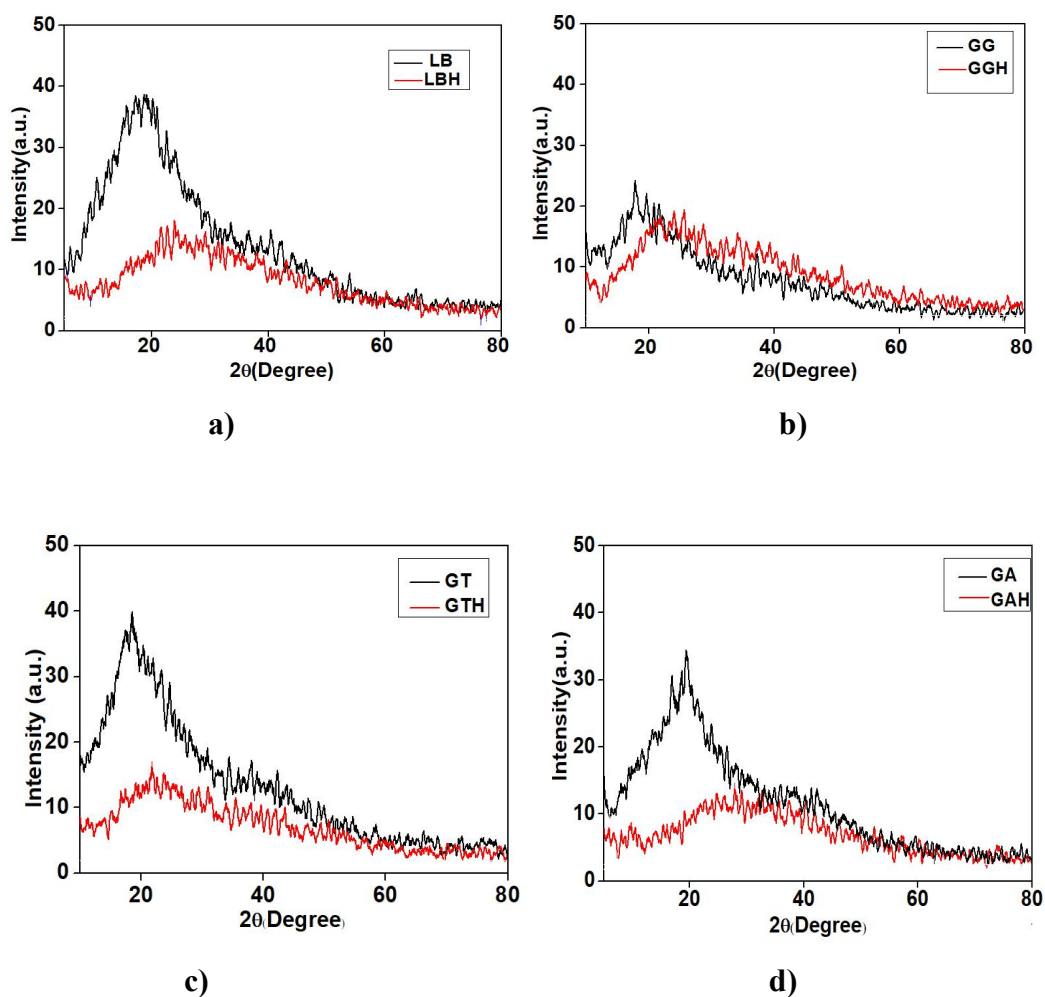
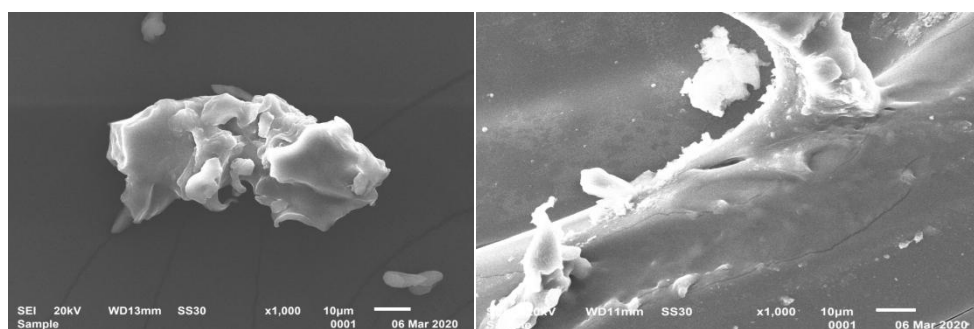


Fig.25. Representation of XRD plot a) LB & LBH b) GG & GGH c) GT & GTH d) GA & GAH.

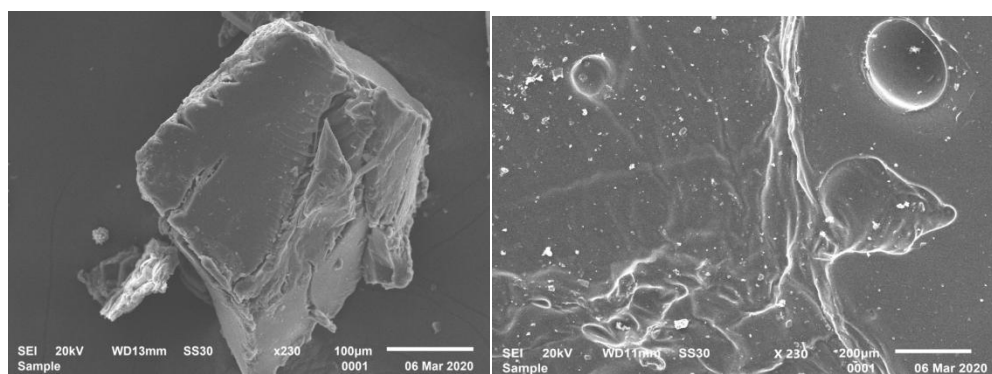
4.3.4 Scanning Electron Microscopy

SEM analysis of each hydrogel (LBH, GGH, GTH, GAH) and their gum (LB, GG, GA, GT) has been taken and is presented in Fig.26. As shown in the figure super absorbent hydrogels were having a porous morphology with a rough surface which seemed to be responsible for its swelling behaviour as it allowed more liquid to penetrate in these voids. These pores were associated with higher swelling as they allowed direct diffusion of liquid in the crosslinked network. The Gums were compact and having a non-porous surface which defines that Gum has been modified, forming interpenetrating networks due to the presence of cross-linking in them.



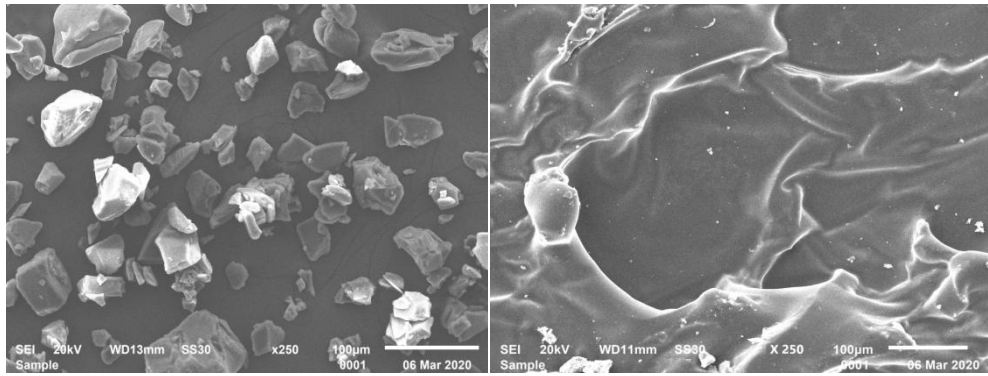
a)

b)



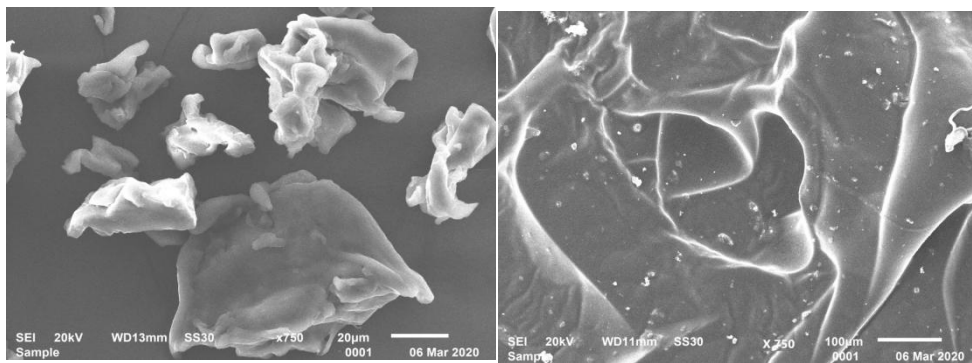
c)

d)



e)

f)



g)

h)

Fig.26. Surface morphology of each gum with its respective hydrogel a) LB b) LBH c) GG d) GGH e) GT f) GTH g) GA h) GAH

4.3.5 Solid-State NMR

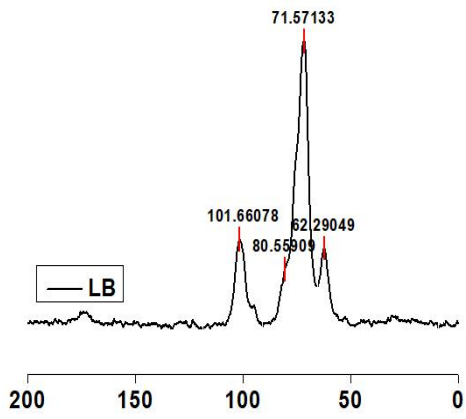
Solid-state ^{13}C NMR of gum and their hydrogel is depicted below in Fig.27. For the LB NMR spectrum, a peak was observed at $\delta = 101.66$ ppm, which was due to the mannose unit of C_1 of LB. The Signal peak at $\delta = 80.55$ ppm observed which represented the C_4 mannan whereas C_6 Carbon of LB mannose unit observed a peak at $\delta = 62.29$ ppm [131]. The hydrogel synthesized i.e. LBH showed a new signal peak at $\delta = 181.47$ ppm due to the carboxylate group ($-\text{COO}$) of poly(sodium acrylate). The signal peak at $\delta = 59.45$ ppm was for the C_6 mannan carbon atom of gum. Lastly, the peak at $\delta = 41.97$ ppm showed the presence of methylene carbon which was due to crosslinker present in it [132]. The peaks clearly showed the modification of gum in hydrogels. For GG NMR spectrum, a peak observed at $\delta = 175.51$ ppm due to the presence of acid D-glucuronic acid, peak at $\delta = 104.20$ ppm due to presence of cyclic

saccharides in its chain, peak at $\delta = 64.43$ ppm due to existence of $-\text{CH}_2\text{-OH}$ group in its chain which confirmed the structure of gum [133]. For GGH, the peaks observed at $\delta = 181.05$ ppm due to $-\text{COO}$ group present due to poly (sodium acrylate). The peaks at $\delta = 104.39$ ppm, $\delta = 80.26$ ppm & $\delta = 60.04$ ppm confirmed hydrogel synthesis. The new peak at $\delta = 42.36$ ppm was due to the introduction of crosslinks in the hydrogel due to N, N-MBA. NMR spectrum of TG, a peak at $\delta = 104.10$ ppm due to cyclic saccharides present in its main chain, peak at $\delta = 81.53$ ppm for a C_3 carbon atom and a peak $\delta = 63.07$ depicted $-\text{CH}_2\text{-OH}$ group in the natural gum [134]. Similarly, NMR peaks of TGH at $\delta = 181.37$ ppm, $\delta = 60.53$ ppm, $\delta = 42.06$ ppm confirmed the synthesis of the hydrogel by crosslinking poly (sodium acrylate) on TG with the help of N, N-MBA. For GA, a peak observed at $\delta = 176.20$ ppm for $-\text{COOH}$ group present in its backbone, peak at $\delta = 104.20$ ppm for the presence of pyranose cyclic ring of an anomeric carbon atom of GA. A signal at $\delta = 72.25$ ppm depicted $(-\text{CH-OH})$ group and a peak $\delta = 62.75$ ppm due to CH_2OH present in the gum [128]. In the crosslinked hydrogel i.e. GAH the peak was observed at $\delta = 181.47$ ppm, which was due to the presence of poly(sodium acrylate). The peaks at $\delta = 91.91$ ppm & $\delta = 81.73$ ppm confirmed the structure of GA present in hydrogel. The introduction of a new peak was observed at $\delta = 41.28$ ppm, which was due to N, N-MBA. Hence, based on the ^{13}C NMR data it is quite evident that all gums have been converted into super absorbent hydrogels in each case. The tabular form is represented below in Table 9.

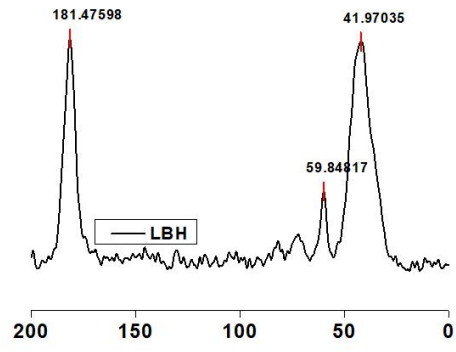
Table 9: Represents the NMR peaks for all hydrogels with their chemical shifts along with functional groups.

Hydrogels	Chemical shifts (δ in ppm)	Functional groups	References
LBH	181.475 ppm 59.84 ppm 41.97 ppm	Due to $(-\text{COO})$ of poly(sodium acrylate) Due to C_6 mannan carbon atom of gum presence of methylene carbon due to crosslinker.	[132]

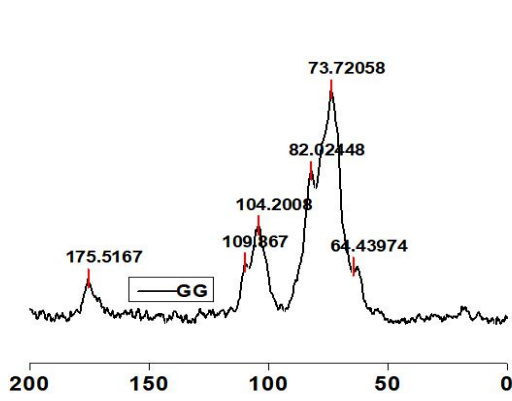
GGH	181.05 ppm 104.39 ppm 60.04 ppm 42.36 ppm	Due to (-COO) of poly(sodium acrylate) Presence of cyclic saccharides in gum. Existence of -CH ₂ -OH group in the gum Introduction of crosslinks	[133]
TGH	181.37 ppm 42.06 ppm	Due to (-COO) of poly(sodium acrylate) Due to crosslinker i.e. N, N-MBA	[134]
GAH	181.47 ppm 91.91 ppm 81.73 ppm 41.28 ppm	Due to the presence of poly(sodium acrylate). presence of pyranose cyclic ring of an anomeric carbon atom of the gum. (-CH-OH) group present in the gum. Due to N, N-MBA which is used as crosslinker.	[128]



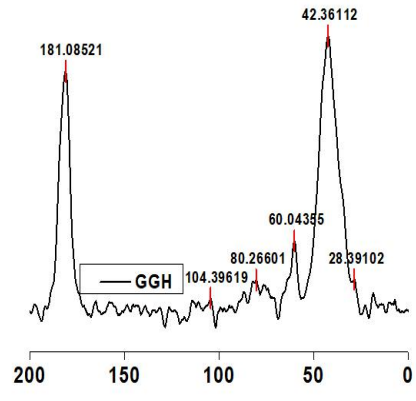
a)



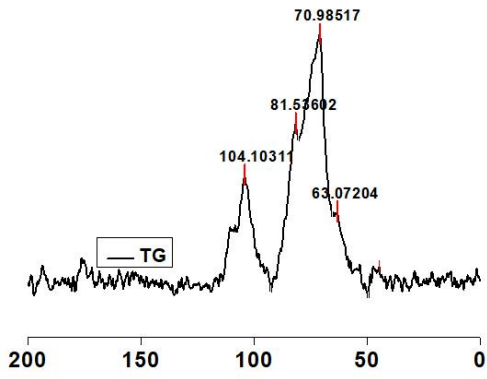
b)



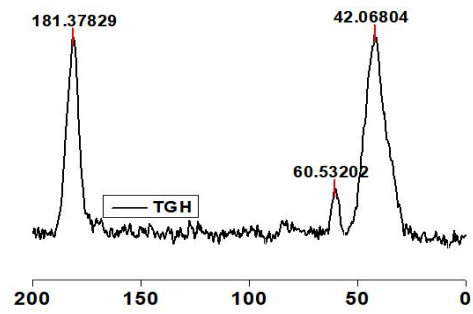
c)



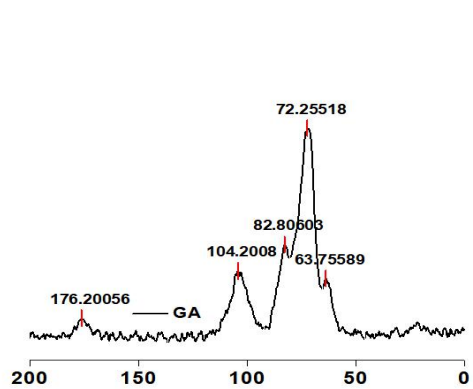
d)



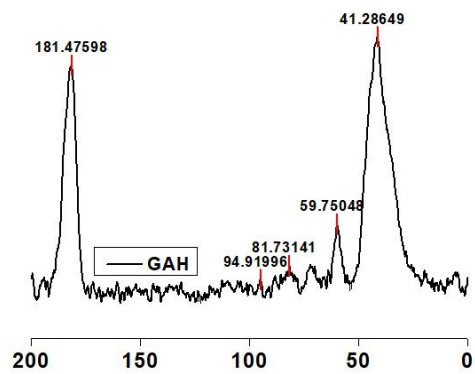
e)



f)



g)



h)

Fig.27. ¹³C NMR Spectrum of a) LB b) LBH c) GG d) GGH e) TG f) TGH g) GA h) GAH

5. Conclusion

Natural Gum based hydrogels have shown their major utility in many areas due to their bio-compatible, toxic, and eco-friendly nature. The produced hydrogels along with their gum were analyzed using TGA, SEM, XRD, ^{13}C NMR, and swelling studies, and from this analysis, it is evident that gums have been modified into interpenetrating 3-D Networks. All hydrogels have been synthesized with the help of their native gums, which resulted in a crosslinked structure that can infuse a huge amount of water in its pores. SI was found to be maximum for LBH, in DW followed by GAH, GGH, and GTH respectively as shown in Table 4. Thermal stability also tends to increase as compared to their native gum. GGH, out of all had the highest degradation temperature due to leftover residual mass at 772°C that has a weight loss of 33.01 %, followed by LBH at 31 %, TGH at 30 %, and GAH at 28.41 %.

The FTIR spectrum and ^{13}C NMR helped in revealing the functional groups present in the gum and hydrogel hence confirmed the synthesis of the hydrogel. XRD analysis helped in interpreting the sharp peaks of gums which were due to semi-crystalline nature which was further altered to broad peaks showing amorphous nature due to modification of gums in crosslinked gel in which most of the alteration was seen in GAH that was 27.88° followed by GGH in 24.11° , LBH at 23.88° and TGH at 21.72° . SEM analysis helped in determining pores size, which was visible and helped in depicting surface morphology of gum and hydrogels. All four hydrogels synthesized using natural gums have shown the ability for water absorption due to their pores in their surface which is quite evident from SEM analysis and hence have shown tremendous swelling behaviour in a different buffer solution which makes them potent candidate in wound healing for the absorption of necrotic cell tissue from the wound and incorporation of the drug for healing activity, hence making its potential utility in biomedical, pharmaceutical and drug delivery applications. Hence all the objectives are completed successfully.

6. References

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