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Recent Advancements In Sustainable Hydrogels for Sensing, Temperature Response, and Catalytic Applications

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ABSTRACT

Over the previous few decades, the interest of chemists has shifted toward material chemistry. The main reason behind this philosophical shift is the straitened properties and dearth of wanted materials to fulfill the surfeited demand. Material chemistry develops intellectual thoughts to manipulate the smart materials of promising, desirable characteristics to suit further use. Improvements from proffer solid to polymer and molecular to nanomaterial unfurled its scope towards catalysts, sensors, molecular transporters, and filters with the capability of immense scientific, social, and pecuniary impact. Nowadays nanomaterials found profound exploitation at the industrial, analytical, and domestic levels.

Keywords: Hydrogel, Optical property, Sensing, Catalytic activity, Temperature response.

1. Gel

A gel consists of two constituents, in which some cross-linkers three-dimensionally interconnect the polymer chains. This structurally complex 3D network material (component 1) forms a rigid network, accommodating a relatively large proportion of fluid (component 2). Both inorganic and organic molecules can act as precursors for gelation. Depending upon the

composition of the gel, the forces of interaction for cross-linking may differ from primary interaction to secondary interaction¹⁻³. Gels are mushy materials Seemed solid but are Susceptible to undergoing large contortion in response to external stimuli⁴.

1.1. Polymer gel

Polymer gel consists of a three-dimensional network of

elastic polymer chains including fluid between the interstitial pores of the 3D network⁵. Polymer gel exists in two phases swollen and collapsed, volume transition between these phases can occur continuously or discontinuously⁶. Broadly polymer gel can be classified into two classes based on dispersion medium, Aerogels, and Lyogels. In aerogels, the dispersion medium is gas, while gels containing liquid as a dispersion medium are known as lyogels^{7,8}. Lyogels containing non-polar liquid between their interstitial spaces are called organo-gels, these gels tend to swell in the organic solvent. If interstitial spaces are occupied by water, then they are known as hydrogels⁹.

2. Hydrogel

Hydrogels are a 3D polymer network with a sufficient degree of cross-linking, they are capable of absorbing and retaining a large amount of water or aqueous solution (hundreds to thousands of times of its mass) without dissolution¹⁰. Their degree of flexibility is comparable to that of natural tissue due to their large water-retaining capacity. The water-retaining proficiency of hydrogel arises due to the hydrophilic functional group affixed to the polymeric backbone; their resistance to dissolution is due to cross-linking between the polymer chains¹¹. Nonetheless, hydrogels contain large water content but they show the characteristics of both solids and liquids, the transport of small molecules and ions across the hydrogel by diffusion is analogous to liquids, meanwhile at the macroscopic level, they exhibit the characteristics of solids; preserve their shape and show mechanical strength like solids¹². These Characteristics make them an appropriate candidate for drug delivery, tissue engineering¹³, contact lens¹⁴, sensor¹⁵, and catalysis¹⁶. A mesh is a solid portion of a 3-D network of crosslinked polymer chains as shown in (Figure 1)¹⁷

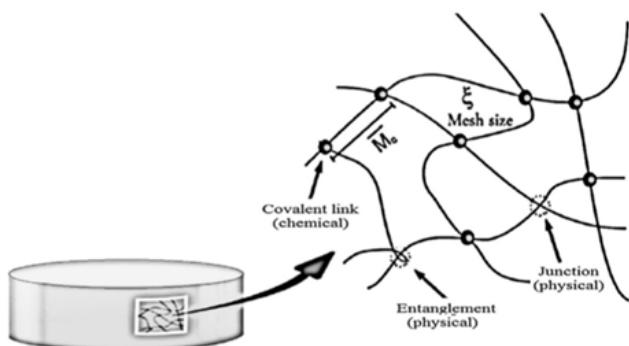


Figure 1: Structural chemistry of a hydrogel, “Hydrogels can be classified as follows”¹¹.

2.1. Hydrogels based on source

On the base of origin, hydrogels are categorized into two groups. Natural hydrogels i.e. polysaccharides (like hyaluronic acid, alginate, cellulose, and chitosan), proteins (e.g. collagen and gelatin), DNA, etc³. Synthetic hydrogels are fabricated by using various types of precursors (one or more than one type)¹⁸. Poly (NIPAM-co-2 HEA)¹⁹, poly (N, N'-diethyl aminoethyl methacrylate), poly (N-isopropyl acrylamide)²⁰, poly (N-isopropyl acrylamide-co-vinyl phosphonic acid)²¹ are some examples of synthetic polymers.

2.2. Hydrogels based on polymeric composition

On the base of the polymeric composition, there are three classes of hydrogels.

2.2.1. Homopolymer gels:

Those hydrogels which are

synthesized by using a single type of monomer are known as homopolymer hydrogels²². The Structural framework depends upon the nature of the monomer, the nature of the cross-linker, and the type of polymerization technique used for its fabrication. Polyethylene and glycol-based hydrogels are examples of homopolymer gels. These types of polymers contain only one type of monomer in their polymeric network, so their properties are limited²³.

2.2.2. Copolymer hydrogels: Hydrogels that are fabricated by using more than one type of monomer are termed copolymeric hydrogels; usually, one type of monomer is hydrophilic. Depending upon the type of precursors and reaction conditions monomers can be arranged to form different configurations (i.e. random, alternate, or block) to form a polymeric network²⁴. Due to their multi-responsive nature, they have several biomedical applications e.g. drug delivery²⁵.

2.2.3. Multi polymer hydrogel: Multi-polymer hydrogels are fabricated by using three or more types of monomers by polymerization technique and cross-linking reaction. Poly (acrylic acid-2-hydroxy ethyl methacrylate)/gelatin hydrogel is an example of a multi-polymer hydrogel. They are multi-responsive due to the presence of more than one type of monomer²⁶.

2.3. Semi-interpenetrating polymeric hydrogels

In these types of hydrogels, one linear polymer penetrates other cross-linked polymeric networks without forming any other chemical bond between them. Due to the absence of an interpenetrating network, they retain the rapid pH and temperature-responsive behavior, but they can adjust their pore size and sluggish drug delivery. One example of this type of gel is the entrapment of poly allyl-ammonium chloride in acrylamide /acrylic acid copolymeric hydrogel. The resulting hydrogel contains both covalent as well as ionic bonds, the covalent bond is responsible for the stability of the 3-D network and the ionic bond is responsible for higher mechanical strength and pH-responsive behavior²⁷. These types of gels have a large no of applications in drug delivery, for dyes, and heavy metal separation²⁸.

2.4. Interpenetrating hydrogels

Such types of hydrogels are synthesized by intimate cross-linking of two independent polymers, at least one component is the cross-linked polymer and the other component is non-crosslinked,^[11] This is fabricated by immersing an already synthesized hydrogel into the solution of the initiator and precursors. The interlocked cross-linked polymeric component is responsible for its stability and morphology. The component polymer of such hydrogels cannot be separated unless the chemical bond between them is broken²⁹. The main advantage of this method is that we can produce stiff and dense hydrogel metrics with desirable physical and mechanical properties. The kinetics of drug delivery can be controlled by tuning the pore size and surface morphology³⁰.

2.5. Classification based on physical appearance

Based on the physical appearance, there are three classes of hydrogel, as shown in (Figure 2)³¹

2.5.1. Solid hydrogels: Those hydrogels that exist in a solid state at room temperature are known as solid hydrogels. They

can shrink and swell in liquids: like water, biological fluids, and buffer solutions. Depending upon the interest in using inorganic metal partials inside these types of hydrogels, they can glow; which gives prominent electrical, magnetic, and optical properties³¹.

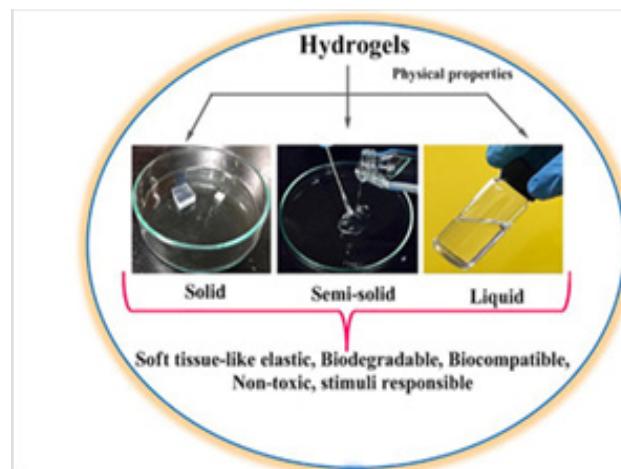


Figure 2: Physical properties of hydrogels.

2.5.2. Semi-solid hydrogels: These types of hydrogels possess strong adhesive interaction with other types of interaction (hydrogen bonding, Vander walls interactions, and electrostatic interaction) and a network of soft tissues. Two different kinds of materials are used to fabricate them. One of these must be natural like plant gum, poly (N vinyl Pyrrolidone), Polycarbophil, Carbopol, etc³². The molecular weight of selecting polymeric material must be ≥ 100 , which enhances the flexibility of hydrogel. Due to bio-adhesive properties, these hydrogels are named bio adhesives³³.

2.5.3. Liquid hydrogels: Such hydrogels are in a liquid state at room temperature, they exist as soft tissue-like elastic material at a specific temperature, and they are easily fabricated³⁴. Due to their self-adjustment in their pore size, in response to the environmental condition, they have a diverse number of applications: e.g. organic and inorganic drug delivery processes. Due to their hydrophilic nature, it is easy to incorporate protein inside them, so they are injected inside living organisms for Vivo applications³⁵.

2.6. Classification based on type of cross-linking

There are three types of classes Based on this classification, physically cross-linked, chemically cross-linked, and Double-Network Hydrogels³⁶.

2.6.1. Reversible or physically cross-linked hydrogels: In these types of hydrogels, polymeric chains interconnect by molecular entanglement or secondary forces: like hydrogen bonding or ionic interactions. The central role in forming chemical interactions is hydrophobic interactions. Depending on the environmental conditions such as pH and temperature, they can dissolve^{37,38}. In recent years, scientists have been interested in preparing physically cross-linked hydrogels. The main advantage of this type of hydrogel is that there is no need to use cross-linking agents. Cross-linking agents do not only affect the integrity of hydrogels but some are sometimes also toxic, therefore, must be removed before further use³⁷.

2.6.2. Permanent or chemically cross-linked hydrogels: In these types of hydrogels, the polymeric chains interconnect by a permanent covalent bond, so they are unable to dissolve. They

are stable³⁹. Depending upon the nature of functional groups present in the 3-D networks, they can be charged or neutral. They may show a change in their shape in response to the external electric field and other stimuli⁴⁰. They have more mechanical Strength and degradation time as compared to physically cross-linked hydrogels³¹.

2.6.3. Double-network hydrogels: In these types of hydrogels, there are two types of cross-linking (both physical and chemical) are present. It overcomes the disadvantages of using either physically cross-linked or chemically cross-linked hydrogels. A recently reported graphene-polymer composite with excellent mechanical properties and self-healing capability is an example of a dual-network hydrogel³⁷.

2.7. Classification based upon stimuli response

Stimuli-responsive hydrogels show an unexpected change in growth action, mechanical strength, and structural network in response to external stimuli: so, they are known as Smart hydrogels, As shown in schematic (Figure 3)⁴¹ Light, Temperature, Pressure, mechanical stress, and intensity of various sources of energy are examples of physical stimuli.

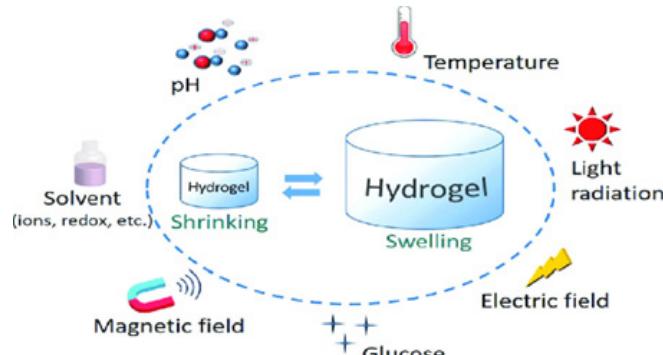


Figure 3: Stimuli-responsive behavior of hydrogels.

These stimuli are responsible for the change in the interaction between the components of gels at specific critical points. Another type is chemical stimuli, including pH, ionic factors, and chemical reagents: they are responsible for the change in the interaction between the solvent and component of gels and within gel at the molecular level⁴². Many multi-responsive hydrogels are also reported, like a polyacrylic acid-co-polyvinyl sulfonic acid-based hydrogel⁴³.

2.7.1. pH-responsive hydrogels: Such polymeric gel, which contains the ionic pendant groups, is pH-responsive. In response to the environmental conditions, these groups pick and lose protons. They show a sudden dramatic change in their ionization (pK_a and pK_b) at a specific pH, so they show sudden volume change at a certain pH. This volume transition occurs due to the generation of the electrostatic force of repulsion between the ionized groups, which creates osmotic pressure for swellings. Both cationic and anionic hydrogel show pH-responsive behavior. Anionic Hydrogels are made up of monomers containing functional groups (like carboxylic, sulphonic, and phosphonic), which donate protons above pK_a , creating the same charge at different sites within the hydrogel. But cationic hydrogels contain functional groups like amines where protonation occurs below pK_b ; this leads to the production of similar charges at different sites within the 3-D network, which is responsible for volume transition⁴⁴. The degree of swelling is controlled by controlling the amount of cross-linker during the preparation of hydrogel, ionic charge, pK_a , or pK_b value of the

ionizable groups, hydrophobicity, hydrophilicity, and degree of ionization⁴⁵.

2.7.2. Temperature-responsive hydrogels: Such hydrogels that can shrink or swell in response to the temperature of surrounding fluids are known as temperature-responsive hydrogels⁴⁶. Chitosan-based copolymer hydrogels extensively studied temperature-responsive hydrogels, like methylcellulose, hydroxypropyl methylcellulose, and NIPAM. Poly (NIPAM) shows a sharp phase transition at a temperature of 34.43 °C in water. P (NIPAM)'s LCT can be altered by the copolymerization of other monomers. It can be increased by copolymerization with hydrophilic monomers and can be decreased by copolymerization with hydrophobic monomers. However, the grafting of such a functional group does not have any appreciable effect on LCT. There are three types of temperature-responsive hydrogels³⁷.

2.7.2.1. Positive temperature hydrogels: They are known as upper critical solution temperature (UCT) hydrogels. They are in a swell state at a temperature above their UCT, but they exist in a shrinking state at a temperature below their UCT. So as the temperature becomes below then the UCT dehydration occurs and they release water from their interstitial pores. At elevated temperatures, the breakage of hydrogen bonds occurs, so this creates the same charge on different sites. As a result, the positive temperature hydrogel exists in the maximum swelling state above its UCT²⁰.

2.7.2.2. Negative temperature hydrogels: Such types of hydrogels exist in the swell state, at a temperature below the LCT; and as the temperature rises above its LCT, it shrinks. The most important parameter for such types of hydrogels is LCT. LCT is controlled in no of ways: by changing the solvent composition, by mixing a small amount of ionic copolymer, or by changing the hydrophobic and hydrophilic co-tenant polymeric hydrogels⁴⁷. Their 3-D network contains both hydrophobic and hydrophilic parts. At a temperature below the LCT, the hydrophilic part interacts with water or fluid by forming a hydrogen bond. Due to solvation, the swelling will increase. While at the temperature above the LCT, the hydrophobic interaction between various parts of the hydrogel becomes dominant; meanwhile, the hydrogen bonding becomes weaker. So, the hydrogel will exist in shrink form at a temperature above its LCT.

2.7.2.3. Thermoreversible hydrogels: Such type of hydrogels has the same kind of composition as that of positive temperature hydrogels and negative temperature hydrogels. However, they differ in their bond type from the previous two types of thermoresponsive hydrogels. Instead of covalently cross-linked polymer chains that show volume change transition, they undergo a sole gel phase transition. Different types of hydrophobic and hydrophilic interactions are responsible for the abrupt change in their volume. In the swollen state, the water molecules organize around the hydrophobic component of the gel (as in the iceberg) by forming a hydrogen bond with the hydrophilic part of the gel. At CST, the hydrophobic and hydrophilic interaction between polymer-polymer chains and solvent-solvent becomes greater than the polymer-solvent interaction. So, they exist in shrink form at CST^{37,48}.

3. Synthesis of hydrogels

Hydrogels can be synthesized either from natural or synthetic polymers. The chemical strength of synthetic polymers is greater, and their degradation process is slow compared to natural

polymers. The greater durability of synthetic polymers is due to their greater mechanical strength. These two properties are optimized during synthesis according to the desired use. Their synthetic techniques are similar to those techniques, which are used for the synthesis of cross-linked polymers. The schematic diagram for the synthesis of hydrogel is shown in (Figure 4)¹¹

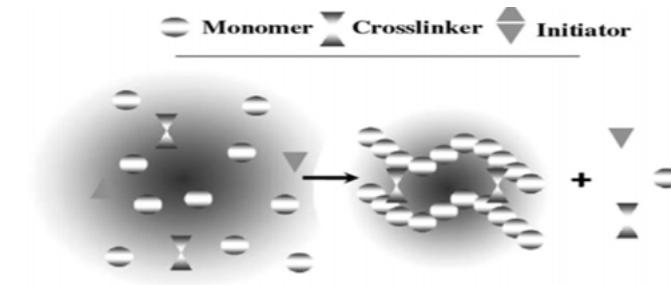


Figure 4: Schematic diagram of hydrogel preparation.

3.1. Bulk polymerization

It is the simplest method of synthesis, in which one more type of monomer polymerizes to give the hydrogel of desired properties. The type of initiators used depends upon the nature of monomers. The rate of bulk polymerization is very high. There is a sharp increase in viscosity as the reaction proceeds, and heat dissipation is a big problem of this technique. These problems can be minimized, by controlling the rate of polymerization. The end product of bulk polymerization is the glassy, hard, and transparent homogenous gel. It swells to form soft and flexible materials when immersed in water.

3.2. Solution polymerization

In solution polymerization, the mixture of (neutral or ionic) monomers and cross-linkers is dissolved in a suitable solvent. The redox initiator is used. The polymerization is initiated either thermally or by irradiating UV light. After completion of the reaction: the unreacted monomers, oligomers, initiator molecules, and other impurities are removed by washing with distal water or DIW. The main advantage of this technique over bulk polymerization is the use of a solvent that acts as a heat sink. The most common solvents used in this technique are water, ethanol, a mixture of water or ethanol, and benzyl alcohol. For the removal of the synthetic solvent, the prepared gel may swell in water.

3.3. Suspension polymerization/Inverse suspension polymerization

The end product of this method of synthesis resulted in the form of powder or beads, so no further grinding is required. “The process in which water-in-oil system (W/O) is used is termed an inverse suspension polymerization, while in suspension polymerization process oil-in-water (O/W) is used.” Suspension polymerization is a commonly used technique. During this process, the homo genus mixture of monomers and initiator molecules is dispersed in the organic phase. The addition of a suspending agent and continuous agitation is necessary because dispersion is not thermodynamically stable.

3.4. Suspension by irradiation

If we have to produce the hydrogel from saturated compounds, the reaction is initiated by irradiating with high energy/ionizing radiations like gamma rays or beams of

electrons. This irradiation is responsible for the production of free radicals on the polymer chain and the radiolysis of water. The hydroxyl radicals (product of radiolysis of water), and small fragments radicals (produced by the hemolytic cleavage of a polymer chain) may attack the polymer chains to produce macro-free radicals. These macro radicals may combine to produce a 3-D cross-linked network system. The main superiority of this technique over the chemically initiated method is the production of relatively pure and initiator-free hydrogels; i.e. poly (vinyl alcohols), Poly (acrylic acid), and poly (ethylene glycol) are cross-linked by this method.

4. Nanotechnology

The concept of nanotechnology was given by Nobel laureate Richard P. Feynman in his famous lecture “There are Plenty of Rooms at the Bottom” in 1960. The word Nano originated from Latin and means Dwarf. With the advancement in nanotechnology, a variety of materials is synthesized. “Nanomaterial is such a substance which has at least one dimension in the nanometer range”. There are many types of classification of nanomaterial⁴⁹.

4.1. Classification based on dimensionality

According to this type of classification; basically, there are four types of nanomaterial: i.e. 0D, 1D, 2D, and 3D. Dimensionality is generally represented by the notation “kD/mn”. The value of k represents the dimensionality of overall materials, while the value of l, m, and n designates the dimensionality of the building blocks of the overall material. The value of k must be equal to or greater than the value of l, m, and n. According to this type of class, there are 36 types of nanomaterials⁵⁰.

4.2. Classification based upon composition

There is another type of classification, “which is according to the chemical composition of constituents⁵¹”.

4.2.1. Metal-based nanomaterials: Metal-based nanomaterials may be pure metal or metal alloys⁵¹. Silver-based nanomaterials have catalytic, diagnostic, conductive, antibacterial, and optical applications. Copper-based partials and their assemblies are used, as catalysts, electrical conductors, thermal conductors, antibacterial agents, and lubricant additives⁵¹. The gold-based nanostructure is exploited for drug delivery systems and specialized alloys, medical testing and cancer detections, dissipation of heat, and fuel cells. Due to their low density and high heat resistance, the nanostructures based on Al-Mg alloy, Mg, Al, and titanium are employed in aerospace technology⁵². Because of its unique magnetic properties, the Fe-Si-B alloy is used for the fabrication of electronic devices⁵³. Iron, Platinum, and palladium nanoparticles are used as catalysts⁵⁴.

4.2.2. Metal oxide-based nanomaterials: Titanium oxide, cerium oxide, and zinc oxide nanoparticles are used as sun screeners in cosmetics products due to filter UV radiation. Nano-silica has good optical absorption ability and photoluminescence and is also used in drug delivery systems. Nano alumina coating acts as an abrasion resister acts as a catalyst and is termly conductive. Depending on the nature of iron oxide, it might be used in the biomedical field, as an ion exchanger, or in the fabrication of electronic structures. The iron oxide nanomaterials may be employed as pigments or might be for the purification of water⁵⁵⁻⁵⁷.

4.2.3. Semiconductor nanomaterials: At the nanoscale, the properties of materials are owned by quantum mechanics⁵⁸.

Nano silicon has applications in solar cells and the biological field and photovoltaic etc. gallium nitride and gallium arsenide at nanoscale use in photovoltaic and in the production of light-emitting diodes.

4.2.4. Nanomaterial including silicates, carbonates, and nitrates: Clay (a nano silicate) can take the shape of a nanotube or nanoplates. Clay may be employed as filler in composites to enhance its mechanical properties, improve electrical conductance, and increase resistance against heat or flame. Nanoparticles of silicon nitrides might be used to decrease the possibility of propagation of the crack. Zeolite at the nanoscale has catalytic applications and may be used in membrane fabrications⁵⁹.

4.2.5. Carbon-based nanomaterials: Carbon-based nanomaterials represent one of the best classes of nanomaterials. Graphene is a one-atom-thick sheet made up of carbon. It has promising applications but its industrial synthesis is under development⁶⁰. Grephen's self-alignment in cylindrical form due to wander wall forces from carbon nanotubes⁶¹. Carbon nanofibers are also folded graphene but without perfect arrangements. Fullerenes are spherical, formed by the folding of graphene. Carbon black is amorphous carbon particles left as a byproduct of incomplete combustion^{62,63}.

4.2.5.1. Nano polymers: Nano capsules and nanospheres are examples of nano polymers. They have an important use in drug delivery⁶⁴.

4.2.5.2. Dendrimers: There are two parts of dendrimers, one is an internal core and the other is the outer branch. The branches can be modified according to requirements. They are important candidates for drug delivery⁶⁵.

4.3. Classification based upon origin

There are two types of nanomaterials, synthetic and natural. Naturally occurring nanomaterials are either synthesized within living bodies or by anthropogenic actions outside the bodies. They are found everywhere: inside the earth's crust, in oceans, on the earth's surface, or in the atmosphere. Human DNA is an excellent example of naturally occurring nanomaterials. Numerous varieties of nanomaterials are synthesized in laboratories and on a commercial scale⁶⁶.

5. Synthesis of Nanomaterials

There are two techniques adopted for the synthesis of nanoparticles, “termed as bottom-up method and top to the down method,” as shown in schematic (Figure 5)⁶⁷. A relatively new technique available only in modern laboratories, “the Bottom to Bottom” approach is also adopted⁵¹.

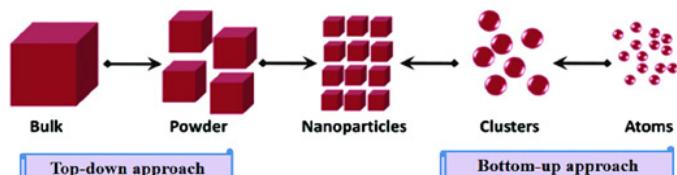


Figure 5: Schematic diagram of top-down and bottom-up synthesis methods.

5.1. Top-down method

In the “top-down method,” we start with a substance in bulk form and end with nanoscale objects by applying either mechanical stress (grinding, milling, or etching); or by using

some chemical agents. The most popular physical method for the top-down approach is lithography, and the other relatively new is electric erosion⁶⁸. It's an easy and cheaper method, but it has many shortcomings; the end product is polydisperse and may have defects in its morphology⁵¹.

5.2. Bottom up method

In this approach of fabrication, we start with atoms or molecules and end with nano-scale objects. During this technique, the first step is the formation of stable nuclei, and the next step is growth. By controlling the growth, we can control the overall size of the resulting nano object. By controlling the growth at different sites, we can control the morphology of results from materials⁶⁹.

6. Hybrid Microgels

A microgel containing metal particles inside their interstitial pores is known as a hybrid microgel. The schematic diagram for the synthesis of hybrid microgel is shown in (Figure 1.6). The nanoparticle is the aggregation of bounded atoms having a three-dimensional radius within the range of a nanometer. The nanoparticles are highly reactive, having a high surface-to-volume ratio and high surface energy. Due to high interfacial activities, they tend to react with contaminants in water, gas, and the earth. So, they are beneficial candidates for controlling environmental pollution. Due to their unstable nature, they have great difficulties in their mass transport. Their separation, reusability, and prevention from agglomeration are difficult tasks. They may be risky for human health and the ecosystem. These risks and difficulties can be minimized; by their incorporation into the gel or other porous material⁷⁰. Such hybrid gels are known as smart materials due to their stimuli-responsive nature⁷¹.

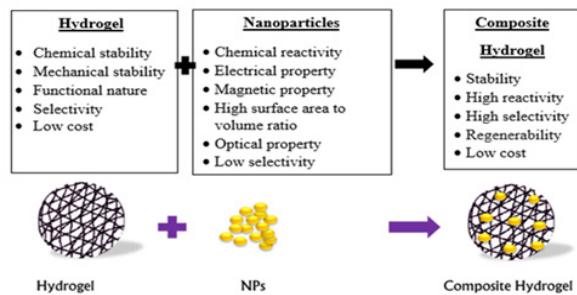


Figure 1.6: Hybrid microgel enhanced physical and chemical properties.

6.1. Responsive hybrid microgels

The hybrid gel may respond to different stimuli (like pH and temperature) depending on its chemical composition. The partial size may vary in response to environmental conditions⁷².

6.1.1. Temperature-responsive hybrid microgels: Depending upon the composition of the microgel, their hydrodynamic volume may increase or decrease with the change of temperature of the medium. The most studied temperature-responsive hybrid gels are poly (NIPAM) and PVC-based. The temperature at which the hybrid system shows abrupt change is known as the VPTT of that hybrid system. NIPAM-based microgel shows a decrease in hydrodynamic radius with an increase in medium temperature.

6.1.2. Glucose-responsive hybrid microgels: Silver NP's incorporated hybrid gel containing Phenyl boronic acid

in their host polymeric gel is glucose-responsive. Poly (3-Acrylamidephenylboronic-co-acrylic acid)-Ag NPs show an increase in hydrodynamic volume in response to a rise in Glucose concentration of the medium⁷³.

6.1.3. pH-responsive hybrid microgels: Such hybrid gel shows the change in hydrodynamic radius in response to the alteration of the medium's pH. Such a system contains ionizable functional groups in the polymeric gel. The hybrid gels containing amino groups in their 3-D polymeric network exhibit an increase in volume due to the protonation of the amino group. So, they exist in a swollen state when the pH of the medium is high. Gels containing carboxylic groups in their 3-D network show an increase in volume at $\text{pH} \geq \text{PKA}$ of ionizing species.

6.1.4. Ionic-responsive hybrid microgels: Such hybrid gels respond to the change in the concentration of ions in the medium. They are responsive to different electrolytes like KCl and CdCl_2 . Investigation shows that VPTT moves towards the lower temperature, as the salt concentration of the medium increases.

6.1.5. Multi-responsive hybrid microgel: Those hybrid gels, which can respond to more than one stimulus, are known as multi-responsive hybrid gels, as shown in (Figure 7).

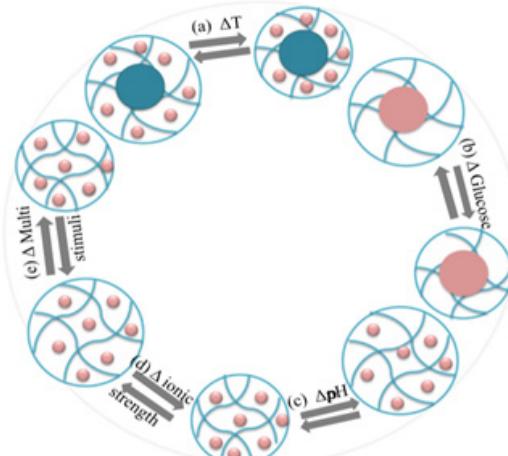


Figure 7: Multi-responsive behavior of silver-embedded microgels towards external stimuli.

They contain more than one type of monomer in their 3-D structure. For example, Ag NPs loaded poly (N-isopropyl acrylamide-co-acrylamide-co-vinyl phenylboronic acid) gel is pH, Glucose concentration, and temperature-responsive. Poly (N-isopropyl acrylamide-co-vinylphenylboronic acid), and poly (acrylamide-co-vinylphenylboronic acid) show sensitivity toward glucose concentration, temperature, and pH. At a given pH of the medium, they show increases in volume with the increase in the concentration of the medium, because more glucose moves inside.

7. In situ synthesis of metal nano particles

For “in situ synthesis,” the ions of metals are loaded inside porous material; porous material is a gel for hybrid gel synthesis. Then the calculated amount of some reducing agent is added for the synthesis of zero-valent metal nanoparticles. The schematic diagram for the Situ synthesis of silver particles is shown in (Figure 8)⁷⁴. Such a metal ion-loaded system may interact with Se^{2-} , OH^{-1} , or S^{2-} depending upon the required composition of the metal nanoparticle^{75,76}.

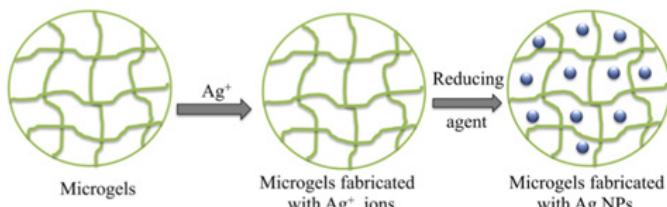


Figure 8: Fabrication of Silver nanoparticles inside microgel by in-situ reduction method.

Other methodologies of in situ synthesis are mixing the nanoparticle and precursor of the host polymeric system. Then the polymerization is preceded by the desired condition to make a hybrid system⁷⁷. The other approach is mixing the precursor of both “metal nanoparticles and host polymer” in a suitable solvent⁷⁸.

8. Silver Embedded Microgels

Silver gives a white and lustrous appearance; these soft transition elements possess excellent catalytic and thermal properties. Silver nanoparticles may be employed as a sensor and as antibacterial agents⁷⁹. They tend to agglomerate because of high interfacial activity. “Microgel provides stability to silver particles due to different anchoring functional groups; moreover, we can tune the size of the particle inside the gel⁸⁰”.

8.1. Applications of silver embedded hybrid microgels

8.1.1. Catalytic applications: Because of the high surface-to-volume ratio, the silver NPs are an efficient catalyst. However, due to their high interfacial activities, smaller NPs aggregate to form bigger partials, so as a result, their catalytic activity decreases. Many substances like dendrimers, surfactants, and polymer brushes are used as a stabilizer for Ag NPs. The silver NPs stabilized by these substances cannot be reused. Moreover, there is a greater chance of poisoning and contamination of catalysts during the catalytic reaction. The Ag NP’s embedded microgel is a very impressive microreactor for catalysis. Their catalytic activity is many times faster than other systems. The inert polymeric network eases the diffusion of reactants toward the Ag NP’s surface and allows the outward diffusions of the product. Their catalytic activity can be tuned by changing external stimuli.

8.1.2. Biomedical applications: Some stimuli-responsive Ag NPs encapsulated hydrogels are used for blood glucose detection and controlled delivery of insulin. The fluorescence intensity of some biomolecules is very low, so there is a problem with their detection by fluorescence analysis. The coupling of such molecules with Ag NP’s loaded microgel can enhance their fluorescence intensity. The detection of porphyrin by metal-enhanced fluorescence effect is reported in the literature. It is used for cell imaging.

8.1.3. Applications in semiconductor devices: When Ag NP’s embedded hybrid gel is encapsulated in a hydrophobic shell, a such hybrid gel is no more sensitive to stimuli except light. Hybrid gel exists in a collapsed state at high temperatures, so the distance between the NPs decreased. When light is irradiated, the reflectance increases because there are no spaces through which light can pass. Their suspension may be used as photonic crystals in different fields.

8.1.4. Calorimetric sensing: In clinical procedures, pharmaceutical industries, food industries, and environmental

analysis, hydrogen peroxide determination is very important⁸¹. Several methodologies like the chromatographic technique⁸², titrimetric analysis⁸³, chemiluminescence⁸⁴, fluorometry⁸⁵, Spectrophotometric analysis⁸⁶, and electrochemistry⁸⁷ are used for (hydrogen peroxide) analysis. These conventional methods are usually time-consuming, difficult, and complicated. With the advancement of nanotechnology, the most commonly used electrochemistry technique is improved by replacing enzyme-based biosensors with nanoparticle-based sensors. Because of efficient catalysis, excellent surface-enhanced Raman scattering, low toxicity, and biocompatibility the Ag NPs earned significant fame in hydrogen peroxide sensing^{88,89}. The silver-embedded three-dimensional network system is an efficient and cheap candidate for hydrogen peroxide sensing⁹⁰.

8.1.5. Optical properties: In response to external stimuli, the hybrid gel can swell and de-swell. As a result, a change in surrounding the effect of solvent, refractive index of solvent, interparticle distance of embedded particle distance, and surface effect changes. So, by changing the condition of the surrounding medium, we can change the surface Plasmon band⁹¹.

9. Conclusion

Recent advancements in sustainable hydrogels have significantly expanded their potential in sensing, temperature-responsive systems, and catalytic applications. Innovations in biodegradable polymers, bio-based materials, and green synthesis methods have improved hydrogel performance while minimizing environmental impact. These materials now exhibit enhanced sensitivity, tunable responsiveness, and improved catalytic efficiency, making them highly attractive for diverse applications, including biosensing, drug delivery, and environmental remediation. Despite these achievements, challenges such as scalability, long-term stability, and cost-effective production remain. Future research should optimize hydrogel properties through advanced material design and integrate them with emerging technologies such as nanomaterials and smart devices. By addressing these challenges, sustainable hydrogels will continue to play a crucial role in developing eco-friendly and high-performance functional materials.

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11. Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

12. Data availability

Data will be made available on request.

13. References

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