

CHAPTER 1

Introduction

Highlights

This chapter provides a concise overview of the research background and the underlying motivation behind the current investigation, along with a brief literature review on the development of organogels. Organogels are briefly described, including their different classifications and the crosslinking process involved, as well as their inherent properties. The mechanistic pathways for formulating organogels and their potential applications in addressing environmental concerns are highlighted. The chapter concludes with a discussion on the objectives and outline of the present study.

1.1 POLYMERS

Marking a significant milestone in research on polymers, the year 2020 commemorates a century of suggested ideology of the term *Makromoleküle* by Noble Laureate Herman Staudinger [1]. These macromolecules of high molecular weight in their repeating structural units exist as polymers. Over a century, intensive research and development efforts have rendered polymers one of the most significant classes of materials in the realm of technology [2]. These versatile materials have been instrumental in the success of several industries, including aerospace, medicine, and electronics, to name a few [3]. As a result, polymers have become a crucial component in modern-day society, contributing to advancements in various fields and offering novel solutions to complex problems. Due to their versatility and practical uses, polymers have become ubiquitous in contemporary society and are extensively utilized across multiple facets, as depicted in Figure 1.1. The adaptability of polymers and its composites has facilitated their substitution for conventional materials in numerous applications, offering an economical and long-lasting resolution for a broad spectrum of requirements [4].



Figure 1.1 Polymers finding applicability in everyday life

Recent advancements in polymers have resulted in the synthesis of a novel two-dimensional polymeric substance by a team of researchers at MIT, USA. The material demonstrates exceptional strength akin to steel, yet retains a weight similar to that of plastic [5]. Moreover, it displays self-assembling characteristics, potentially heralding novel avenues in the realms of manufacturing and engineering.

Polymers can be classified into three states solid, liquid, and gel based on their molecular structure. Solid polymers typically present as amorphous glasses, exhibit a higher level of aggregation. However, they can crystallize like fibers when their chain structure exhibits some degree of order [6]. While polymers are seldom encountered in a liquid state, they often possess a rubbery texture that thickens as the temperature rises. An intermediate state between solid and liquid exists as gels, where solvent molecules permeate among the closely packed network occupying the interstitial places exhibiting remarkable properties of both solid and liquid.

1.1.1 Polymeric Gels

The human way of life has undergone significant transformations with the advent of a pliable "semi-solid viscous substance." This intermediary state, resulting from the combination of solid and liquid substances, characterized by the immobilization of the liquid within a three-dimensional networked structure created by the solid component, is commonly referred to as a "gel" [7]. Throughout the past century and a half, the nomenclature in this field has evolved following the investigations of Scottish scientist Thomas Graham into gels [8]. In 2007, the International Union of Pure and Applied Chemistry (IUPAC) formulated a definitive definition of gels as a "*non-fluid colloidal network or polymer network that is uniformly spread throughout its entire volume by a fluid*" [9]. Gels hold a crucial role in the daily routines of households. For instance, waking up in the morning to brush the teeth requires the usage of gels in the form of toothpaste [10]. Furthermore, gels are extensively employed in the formulation of oral and dermal drug products, such as microcapsules and ointments [11], which exist as oil/water emulsions and oil-based organogels, serving as soft materials for decades. Moreover, these gels are commonly utilized as implants for in situ parenteral administration, where the active substance shifts from a solution to a gel state as the temperature decreases within the body [12].

Formation of a three-dimensional network structure by interconnecting molecules; these networks aided by a liquid medium give rise to gels (Figure 1.2) [13]. The gels exhibit the property to retain solvent within them apart from possessing porous structure with excellent strength due to the presence of network structure [14].

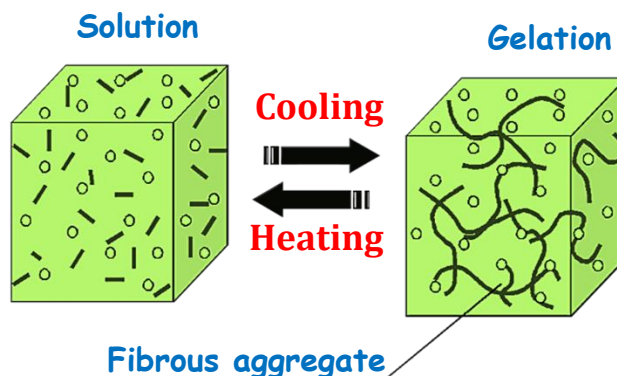


Figure 1.2 Gelation behaviour obtained from solution phase

Polymer gels have garnered significant interest owing to their traditional application in a diverse range of fields. Their distinctive characteristics such as stability, flexibility, and exceptional textural qualities make them valuable components in pharmaceuticals and cosmetics, where they are utilized in formulations such as drugs and lotions [15]. Moreover, their inherent self-assembly properties have led to their extensive application in industries for purposes like adhesion and coating, while their soft structure has also found a place in the food industry, particularly in products like jelly candies (Figure 1.3) [16].



Figure 1.3 Products made up of gels used in daily life

Based on the composition of the solvent of choice, gels can be classified into various categories such as hydrogels, organogels, aerogels, xerogels, and cryogels [17]. A hydrogel is formed when gelators create a three-dimensional network in the presence of water as the solvent, whereas organogels are formed when organic solvents are utilized. Aerogels are produced by replacing the dispersed solvent phase with a gaseous

substance, maintaining the structural integrity [18]. Xerogels and cryogels are obtained through thermal drying and freeze-drying processes, respectively, thereby eliminating the solvent from the network [19]. A detailed overview of polymer gels is provided in Figure 1.4, outlining their classification based on composition, origins, types, and crosslinking methods. The present thesis centers around the development of organogels and their potential applications in the field of pollutant eradication.

Classification of Polymer Gels

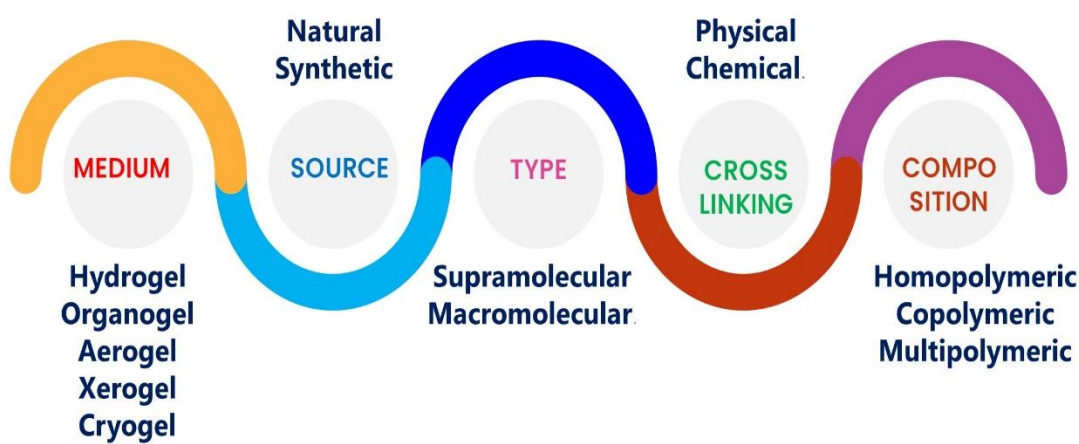


Figure 1.4 Classification chart of polymer gels

1.2 ORGANOGELS

The immobilization of organic solvent within a continuous three-dimensional network structure formed by the self-assembling of gelator molecules comprises an organogel [20]. The three-dimensional network established within the gel structure imparts specific characteristics and valuable insights into the organogel. The interaction between the gelator molecules and the organic solvent triggers self-assembly processes leading to gel formation [21,22]. The network structure effectively prevents the solvent from escaping the structure, thereby ensuring deformability and flexibility. Given that organogels are derived from organic solvents, they can absorb significant quantities of organic solvents or oils while maintaining their gel properties [22]. This property also renders them suitable for accommodating hydrophobic or lipophilic substances, thus enabling compatibility with a diverse array of organic compounds. Due to the cross-linked structure that forms in the presence of an organic solvent, the stability of organogels is significantly increased in a non-aqueous environment when compared to hydrogels. This characteristic renders the gel well-suited for exposure to water [23].

The synthesis of organogels entails a diverse array of gelators, encompassing both low molecular weight organogelators (LMWGs) and high molecular weight organogelators (polymeric), combined with apolar or non-polar organic solvents. The gelator particles interact with the organic solvent to create a network structure through covalent bonding or physical entanglement [24]. Moreover, the gelators also engage in chemical interactions or self-assembly processes to create an interconnected crosslinked network structure. This network architecture facilitates the absorption of an external solvent into the matrix, leading to solvent-gel interactions that hinder its flow towards the outer surface. The external solvent infiltrates the void spaces within the network or the central region of the available pores [25]. The polymer volume fraction parameter was used to evaluate the swelling behavior in the organogel. The extent of swelling from the initial state to equilibrium was measured by calculating the reference/swelling ratio [26].

Being the first of its kind of gel to form, hydrogels can be termed traditional gels [27], widely utilized in various fields. They are predominantly formed using water as the solvent of choice. Despite their popularity, it is important to recognize certain limitations linked to hydrogels, like their inability to accommodate lipophilic drug molecules, and hydrophobic substances, or address oil spillages. In light of these challenges, the concept of organogels was introduced as an innovative alternative to traditional gels.

1.2.1 Types of Organogel

The formation of molecular aggregation within an organogel can be categorized into three distinct classifications based on the interactions and driving forces involved. These classifications are physical organogels, chemical organogels, and hybrid organogels. Understanding the properties and characteristics exhibited by each type of organogel is crucial for advancing and creating materials with specific properties and functionalities. A detailed description of these categories has been provided in the following sections.

(a) Physical Organogels

The formation of a three-dimensional cross-linking network through non-covalent bonding involves various interactions, such as H-bonding, van der Waals forces, π - π stacking, dipole-dipole interactions, and donor-acceptor interactions [24]. The

interactions described are instrumental in the formation of physical organogels [28] and serve as the foundational framework for the subsequent discussions on absorption and adsorption processes in the forthcoming chapters. It is important to note that these gels are relatively unstable since they can undergo a reversible transition from a gel to a sol state when there are changes in environmental conditions, such as temperature, ionic strength, or pH [29]. During these transitions, the three-dimensional network structure of the gel collapses and reforms once the original conditions are restored [30].

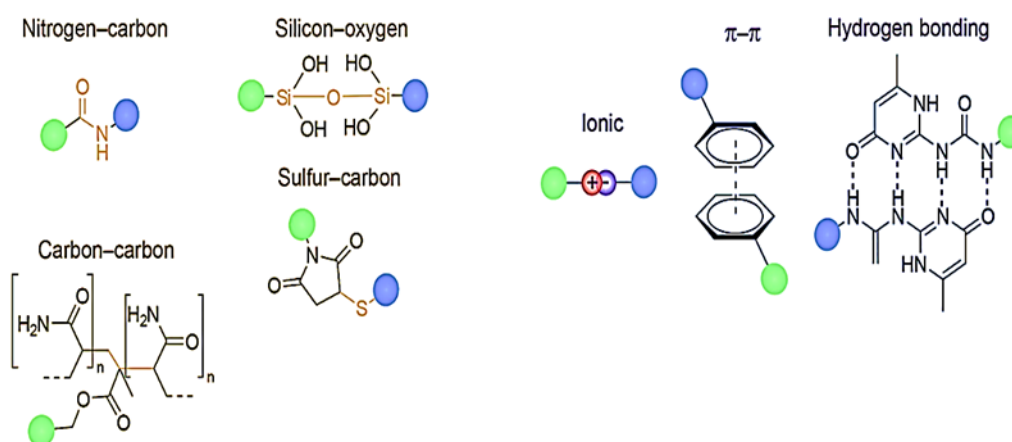


Figure 1.5 Representation of physical crosslinking through hydrogen bonding, π - π stacking, ionic bonding as well as permanent crosslinking through the formation of N-C, S-C, Si-O, C-C bonding [31]

(b) Chemical Organogels

Gels formed through covalent cross-linking or chemical reactions between gelator molecules are commonly referred to as chemical organogels [32]. Crosslinking can occur via polymerization reactions involving multifunctional monomers or through chemical modification, leading to the development of a crosslinking network capable of trapping solvents [24]. The presence of covalent bonds renders these gels resilient and impervious to physical deformation. This three-dimensional network remains unchanged in the face of external stimuli such as dilution or temperature variations, owing to the establishment of a helical configuration [33].

(c) Hybrid Organogels

The entrapment of nanomaterials within physical or chemical organogels has led to the development of hybrid organogels [24], which possess enhanced properties suitable for a wide range of applications. To obtain these hybrid organogels, nanoparticles are suspended within the organic phase, and then gelator molecules are

added. This mixture is heated above the gel-forming temperature (T_{gel}) and subsequently cooled down [34]. In the case of low molecular weight organogels, the entrapment of gold nanoparticles produces a fibrous network. In another study conducted by Pelever and colleagues, the inclusion of multi-walled carbon nanotubes as inorganic nanoparticles was found to increase the thermal and mechanical stability of the gel [35].

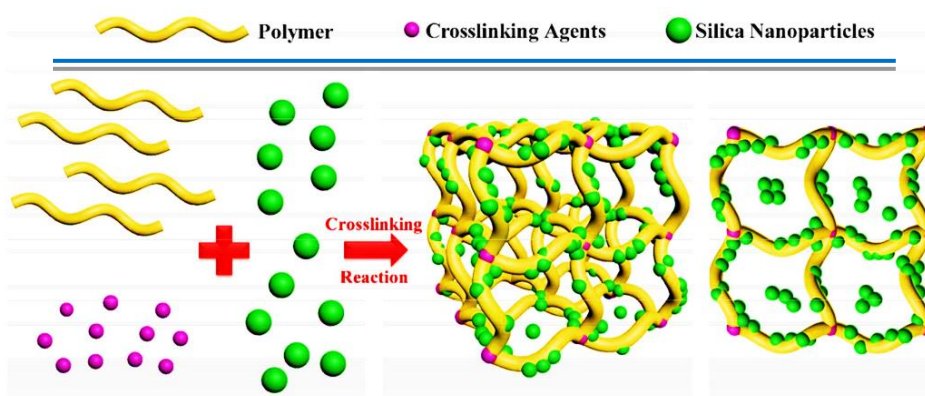


Figure 1.6 Formation of hybrid crosslinking in polymeric network [36]

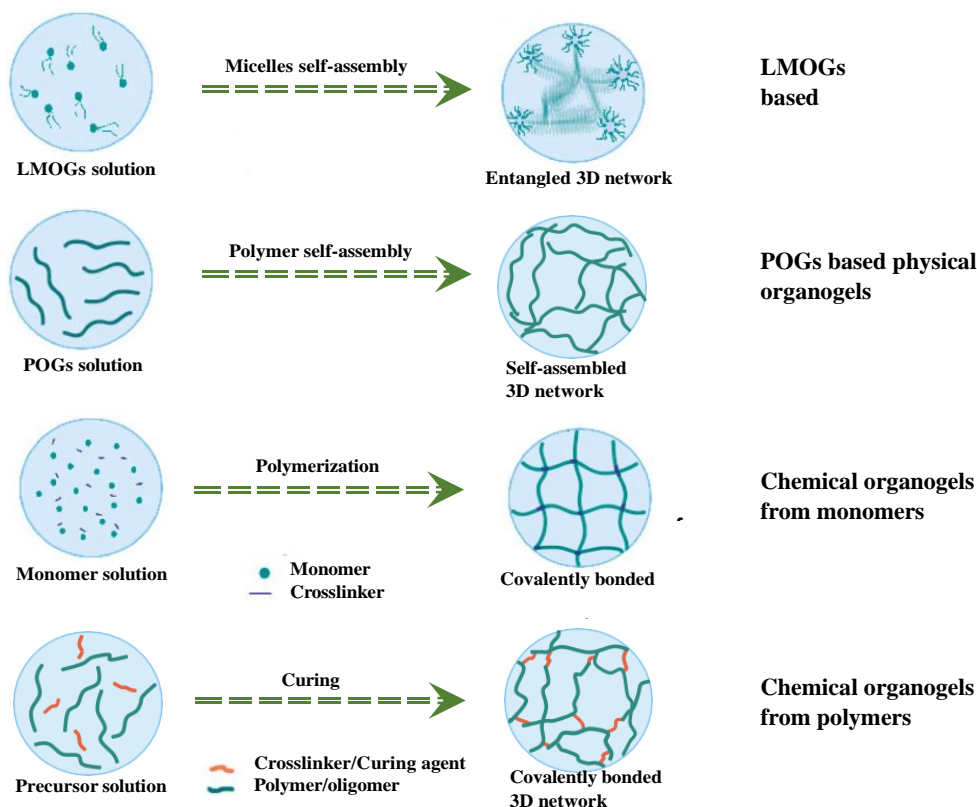


Figure 1.7 Illustrated scheme of gelation mechanism involved in the formation of physical and chemical organogels [32]

An intriguing and distinct progression can be also observed from the solution of organogelators to the construction of network structures, which visually explain the

production of organogels. The successful gelation in a specific solvent depends on achieving a delicate balance between the interactions of the gelator and the solvent aggregate [37]. As organogels exist as chemical and physical, or supramolecular gels, chemical gels are formed by incorporating gelators with high molecular weight or polymers, whereas physical gels employ gelators with low molecular weight. It is of utmost importance to distinguish between them, as they have profound implications for the stability and characteristics of the resultant organogels. The presented figure 1.7 showcases the gelation process involved in the formation of organogels through various network configurations.

1.2.2 Crosslinking Involved in Organogel

The formation of a gel network takes place when molecular chains combine to form a three-dimensional mesh structure through crosslinking, either in the presence of an organic solvent or in an aqueous medium [38]. This crosslinking process results in the formation of physical and chemical gels, based on the type of interaction and bonding involved, which will be discussed in detail below.

1.2.2.1 Crosslinking forming physical gels

Interactions mediated by noncovalent bonding are relatively weak and can be easily reversed [39]. However, the presence of various types of interactions, such as hydrogen bonding, hydrophobic association [40], ionic interactions, and crystalline formations, allows for the formation of temporary networks that give rise to reversible gels [41] with enhanced properties, including increased strength, self-healing ability, and toughness through physical interactions [42]. A supramolecular gel was prepared by Yang via self-assembly using noncovalent interactions. The resulting gel demonstrated excellent recovery of its viscoelastic properties and exhibited important rheological characteristics [43]. To achieve gels with high strength, scientists have combined hydrogen bonding, hydrophobic association, and interactions involving electrostatic forces and van der Waals forces to create gels with remarkable strength [38].

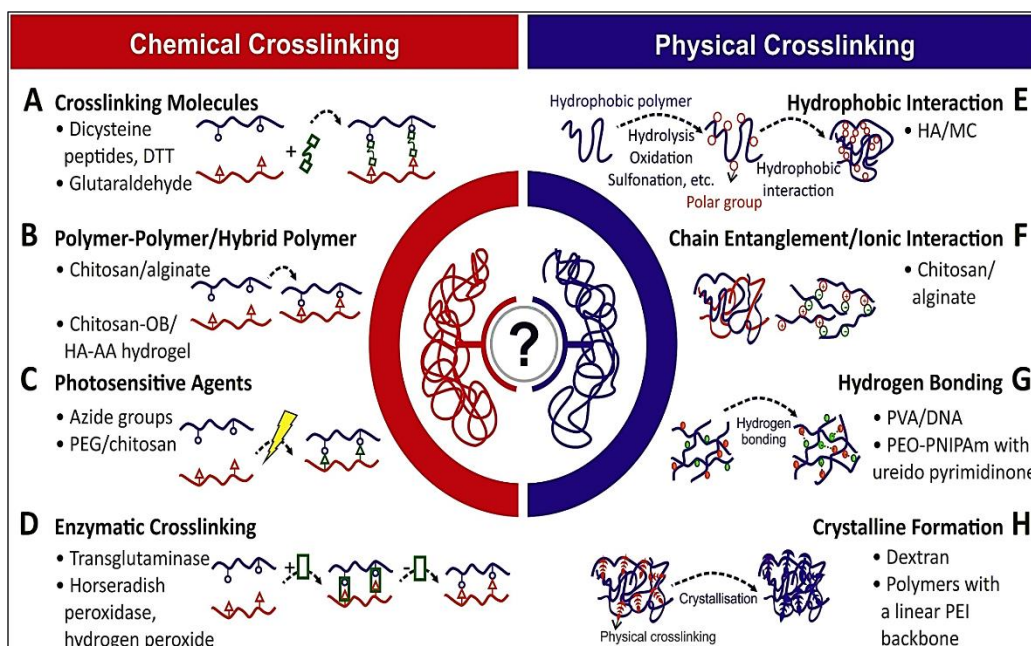


Figure 1.8 Types of methods involved in chemical and physical crosslinking [49]

1.2.2.2 Crosslinking forming chemical gels

The formation of permanent junctions between segments of polymeric chains through covalent bonding results in the formation of irreversible gels [44], which produce strong and impermeable matrices [45]. Crosslinking in such gels occurs based on the composition of the polymeric chain, involving polymerization through addition, condensation, chain growth, and photopolymerization [14]. However, after crosslinking, the structure of the polymeric chain undergoes significant changes in its physical properties [46], including excellent elasticity and the ability to swell in various solvents [15]. A covalently crosslinked organogel, obtained through free radical polymerization between PVA and EG, leads to the formation of a mechanically tough and stretchable double network gel that exhibits anti-freezing properties down to a temperature of $-60\text{ }^{\circ}\text{C}$ [47]. Courtney Fox classified covalently bonded crosslinking junctions into dynamic and supramolecular connections. Dynamic connections remain stable until triggered by external factors, exhibiting self-healing properties. Supramolecular connections involve both covalent and non-covalent interactions, resulting in exceptional durability and strength [48].

1.2.3 Properties of Organogel

Based on the entangled network structure formed, organogels encompass a wide range of characteristics defining their physicochemical properties as listed below:

Depending on concentration and solvent-gelator interactions, organogel exhibits both viscous (flow) and elastic (solid) behavior following the Maxwell model [50]. The property of viscoelasticity can also be validated by the study of stress relaxation [45]. With the increase in shear stress, the network structure of the gel weakens due to the disruption among interacting sites. This grounds the viscous behavior and causes the organogel to flow [51]. As the shear rate is lowered, the organogels behave as solid-like with the gelator molecule assembling to form a three-dimensional network showing elastic properties [52].

Upon heated above the critical temperature, the matrix structure of the low energy thermostable organogel gets disrupted. Interactions among the gelator molecules in the formed network undergo physical disruption due to this addition of thermal energy [24]. However, as the temperature reduces, the coherent interaction between the gelator molecules reposes to its thermodynamically stable configuration. This is where the gel exhibits thixotropically [53]. Figure 1.9 illustrates the thermoreversible and photoresponsive properties of the Azo-mLCP organogel. When exposed to both light as well as temperatures above the sol-gel temperature, the gel transitions into a liquid state, which subsequently becomes stable upon cooling [54]. This thermoreversible property is mainly observed in physical organogels (supramolecular gels) [55].



Figure 1.9 Thermal and photoresponsive property exhibited by the Azo-mLCP organogel [54]

Organogel possesses intrinsic thermostable qualities. The gelator molecules self-assemble to form organogels under specific conditions [50]. This self-assembling renders a decrease in the overall free energy of the system, thus producing thermostable organogel with less energy [56]. Due to this thermostable nature of organogels, they possess longer shelf life which makes them effective for commercial viability [57]. Figure 1.10 describes the formation of organogel using water and ethylene glycol as binary solvents capable of retaining compression and bending at $-40\text{ }^{\circ}\text{C}$ [58]. Thus, organogels are preferred as carriers for use in the cosmetic industry or delivery of bioactive agents.

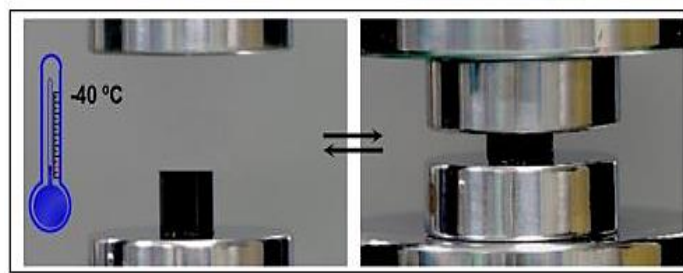


Figure 1.10 Conductive self-healing organogels with stable strain sensitivity at subzero temperatures [58]

With the commencement of the formation of organogel, certain components used rendered compatibility issues in the organogel [24]. With progress in recent years, organogels have been formulated with components like vegetable oils, mango butter, lecithin, and hydroxystearic acid keeping the issue of biocompatibility for its specific use in the pharmaceutical and bio-medical field [59,60]. The photodegradable property of poly(methacrylate) organogels can be visible with the usage of UV light [61], as seen in Figure 1.11.

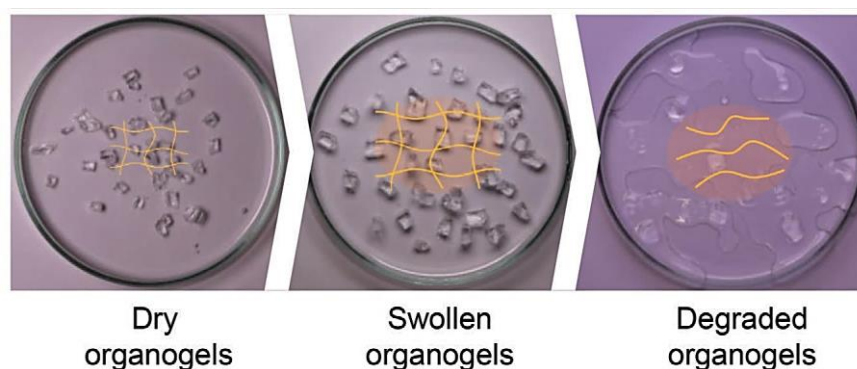


Figure 1.11 Organogels exhibiting biodegradable properties under ultraviolet radiation [61]

Further introduction of bioresources also portrays degradable qualities in the organogel. The degradation rate can be amplified with variations in the concentration of components. Certain physical and chemical stimuli even obligate the degradation of gel solvents.

1.2.4 Factors Affecting the Formation of Organogels

The strategy for the formation of gels depends on the potentiality of the gelator molecules to interact with solvents for gelling [62]. The prime aspect accountable for forming organogels lies with the organogelators as well as the presence of organic solvents. A heated homogeneous gelator solution when cooled down below room temperature often forms gelation. The solvents become immobilized within the fibrous

network, rendering them mechanically inert [63]. Both temperature and concentration play crucial roles in this process. The specific factors responsible for gel formation are elaborated upon below.

1.2.4.1 Solvents

The selection of organic solvents affects gel formation, as the reactivity ratio of the co-monomer for polymerization differs with varying solvent system [64]. The dissolution of the organogelator in the presence of organic solvent is a required prerequisite for organogelation [63]. Hence the presence of solvent plays a pivotal role in the gelation procedure. All sorts of organic solvents along with vegetable oil [65] can be used as solvents for the formation of organogel. However, varying solvent properties may affect the structure of the formed gel as solvent assists in self-assembling and favors interaction involving inter-gelators [66]. The changes observed due to interactions between solvent and gelator molecules were studied by Henry's law constants, Hansen solubility, Kamlet-Taft parameter [67]. Obtaining critical gel concentration is another parameter to determine the self-assembling mechanism of solvents.

As studied by Hu and the group, the decrease in the alkyl chain of the gelator polarity rises, increasing the precipitation. Due to this, the polarity of the solvent may affect the interactions inducing gel formation such as vander Waals forces and hydrogen bonding. Accordingly, solvents with low dipole moment ($D < 1$) as toluene, benzene, xylene, hexane, heptane, and cyclohexane initiate gelation [67]. However, an exception among the solvents was observed as an apolar solvent, dimethyl sulfoxide ($D = 1.04$) was found to gelate all kinds of gelators possibly due to its characteristics of hydrogen bonding. The role of polar solvent during gelation as a cosolvent in supramolecular organogels is to stabilize the system by forming hydrogen bonding, thus causing changes in the structural transformation from gel to crystals [68]. Interactions between gelator molecules and solvent govern the process of gelation as well as the formation of the structural network of the gel [66]. Low interaction of a gelator with solvent (ethyl acetate) yielded highly entangled thin nanofibers (Figure 1.12 a,b) while the same gelator formed fibers of thick and rigid nature (Figure 1.12 c,d) as there was an increase in the interaction between gelator and solvent (acetonitrile). This reveals the crucial role that the solvent medium plays in determining the properties of organogels.

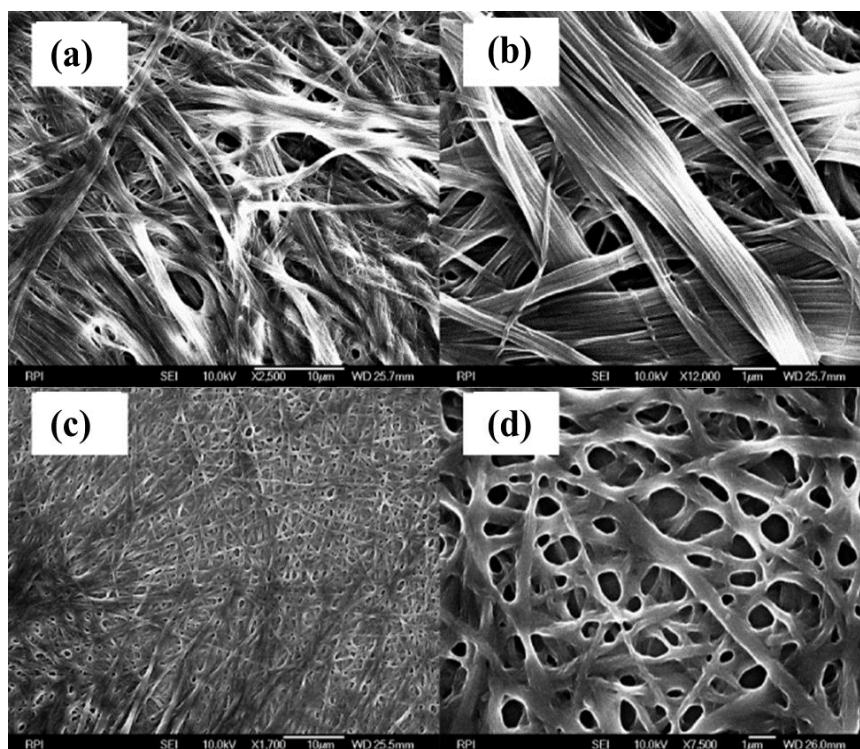


Figure 1.12 Morphological images of organogel formed in ethyl acetate (a,b) and acetonitrile (c,d) with low and high magnification respectively[66]

The congeniality of the solvent with the formed network structure of the gels determines its capacity to swell. However, shrinkage in the swelling gel may occur at times if the boiling point of the solvent used is low [69]. In such cases, the solvent although being entrapped within the three-dimensional structured network, evaporates from the organogel causing shrinkage [70]. Solvents with high boiling points are mostly preferred for maintaining the thermal stability of the organogel [71].

1.2.4.2 Organogelators

Formation of a fibrous network owing to the self-assembling of certain molecules (gelators) immobilizing organic solvents constitutes a three-dimensional gel structure [72]. Organizing these gelator molecules into a fibrous network is generally mediated through interactions based on hydrogen bonding and van der Waals forces as the high directionality and strong bonding probe the behavior of gelation [73]. The self-assembling grounded on these interactions is accountable for a diverse set of organogelators.

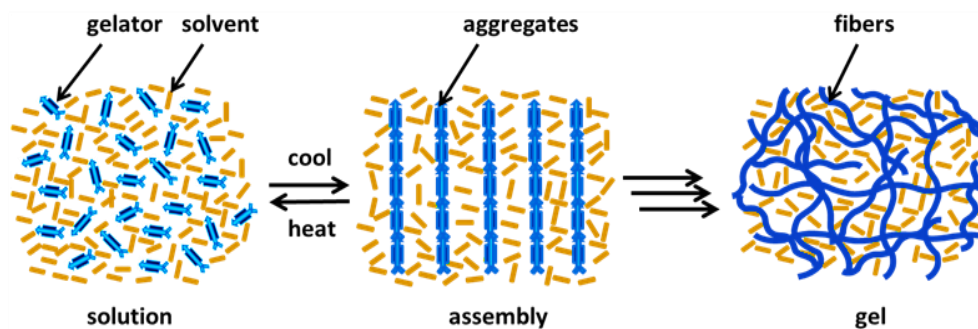


Figure 1.13 Pictorial representation of solvent and gelator assembling to form a three-dimensional network structure[63]

Gelators forming hydrogen bonding include molecules as amino acids, carbohydrates, amides while those with non-hydrogen bonding include steroid based gelators, anthraquinone, tetraline, and anthracene [74]. Assembling of non-hydrogen bonded molecules is mediated through the solvophobic effect as well as π stackings since π - π interaction highly enhances the structural strength [63]. The buildup structure of the organogel along with its properties are determined by the organizing ability of the organogelators as they form bonds by interacting within the molecules [75]. The structure of the gelator also directs the degree of self-assembling in the organogel which resembles either micelle, nanofibers, emulsions, crystal networks, reverse micelle, fibers as well as gel strands [76]. Based on the molecular weight, organogelators can be categorized as low molecular weight organogelators (LMOGs) and polymeric organogelators (POGs), an overview of which is discussed below:

(a) Low molecular weight organogelators (LMOG)

Low molecular weight organogelators are organic molecules with low molecular weight ≤ 3000 Dalton [76]. These gelator molecules interact by forming colloidal arrangements through intermolecular hydrogen bonding, dipole interactions, π - π interactions as well as van der Waal interactions which spread to create three dimensional gel network [77]. These interactions laterally with surface tension play an essential role in sheathing solvents within the organogel [78]. Use of certain organogelators as carbohydrate [79], peptides [80], bis-urea, amides [81,82] involve H-bonding interaction whereas gelators in the form of long chain alkanes often utilize van der Waal interactions which induce a low critical aggregation concentration. The use of certain biological organogelators as cholesterol involves π stacking, non-covalent, and van der Waal interaction during the formation of organogel [83].

(b) Polymeric organogelator (POG)

Polymeric organogelators are molecules with a very high molecular mass, due to which they initiate excellent gelling ability of a wide range of solvents even at a minimal concentration [84]. Modifying the polymeric skeleton may also differ in the capacity of gelation. These gelators simultaneously form supramolecular polymeric (reversible) gels using non-covalent interactions while binding covalently through crosslinking within the three dimensional network forming irreversible gels [76]. Since the crosslinking density for polymeric gelators is considerably low, they are developed effortlessly in contrast to LMOG [24]. The transition temperature while switching from gel to sol state is significantly low and the gels formed have higher strength yielding stable organogels in comparison to other low molecular weight gelators [50]. Conventionally used polymeric gelators include polyethylene, poly(ethylene glycol) [85], polyesters [86], polycarbonate, copolymers of methacrylic acid [87], methyl methacrylate [88].

1.2.4.3 Temperature and Concentration

The stability and formation of the organogels involve the significant role of temperature. With the increase in temperature, there occurs changes in the solubility of the system [89]. This results in an increase in the kinetic energy of the molecules present and a decrease in the formation of intermolecular hydrogen bonding which are the prime causes for self-assimilation [90]. The three-dimensional structure is disrupted and molecules become unstable with the rise in external energy however shelving of the temperature sets up hydrogen bonding within the molecules leading to a well-formed stable structure [91]. Pinhas and group in their work demonstrated that temperature rise enhances the strength of the gel network with the increase in mechanical properties [90]. Temperature changes are subjected to find use in specific applications involving phase transitions [92].

Simultaneously concentration of gelator molecules influences the gelation properties and kinetics of organogel [93]. Rogers and co reported that increasing concentration contended an extremely smooth surface from a greasy surface with a more stiffed texture [94]. Moschakis and group observed a fourfold increase in sterol product as the increase from 5% to 10, 15, and 20% exhibited a well-developed three-dimensional structure with increased strength and hardness as the concentration increases [95]. Further molecular interactions and packing density increased respectively with an

increase in concentration creating a robust matrix structure with improved strength [96]. Concentration also affects the stability of the gel as aggregates are formed using low concentration while higher concentration involves kinetically favorable gels as they rapidly aggregate converting to stable fibrous form even during the initial phase reserving excess solvent forming organogels [97].

1.3 MECHANISM OF ORGANOGELE FORMATION

The process of the emergence of organogels from a self-assembling fibrous network has been fascinating. The interactivity between the gelator molecules and solvent upon heating induced gelation in the matrix substance [24].

Depending on the type of gelator preferred, the formulation of organogel takes place through three general mechanisms. LMOG follows the fluid fiber matrix as well as the solid fiber matrix mechanisms while polymeric gelators observe the polymeric matrix mechanism [76]. Along with it, the traditional method of forming organogel through emulsion can also be taken into account. The primary methods of mechanism differ in the gel system according to its kinetic stability. Assembling of LMOG takes place through intermolecular non-covalent interaction while that of POG through ionic and covalent interaction [50]. Schematic representation of each mechanism is described under respective section heads.

1.3.1 Fluid matrix fiber mechanism

Formation of tubular reverse micelles can be observed in organogel undergoing fluid fiber mechanism [98]. Apolar solution of surfactant on addition of polar solvent observes reverse micelle structure. On further addition of polar solvent, these micelles realign into cylindrical aggregates of mono or bilayers where the polar substance present in the solvent gets incorporated into the micelle structure. These cylindrical aggregates entangle to form a three-dimensional network structure [99]. Due to this change in conformation, organic solvent molecules get trapped in between the mesh spaces leading to an increase in surface tension. Physical organogel (weak gel) or supramolecular gels are formed using this mechanism with a thermoreversible nature [100]. Organogels formed from lecithin and sorbitan monostearate generally exhibit the fluid matrix mechanism. Figure 1.14 represents the fluid matrix pathway.

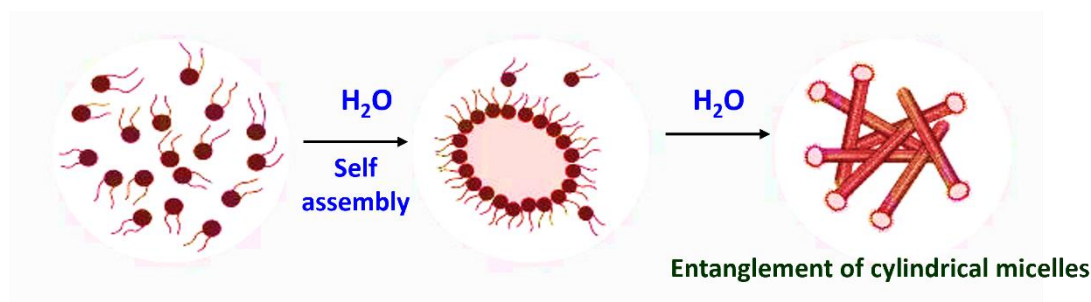


Figure 1.14 Illustrative diagram of organogel formation through fluid matrix mechanism

1.3.2 Solid matrix fiber mechanism

Interaction of the gelator molecules in heated organic solvent gives rise to a solid fiber network [98]. Requiring a minimal concentration (below 15%), the gelator molecules start dissolving with the increase in temperature. With the cooling down of the solution, the dissoluble capacity of the gelator reduces which results in the decrease of the solvent-gelator affinity. As the solution starts precipitating, the molecules self-assemble to form solid fibers upon gelation. The fibers assemble to form solid aggregates by experiencing physical entanglement and crosslinking [76]. The remaining solvent-gelator present stabilizes the system due to which the organogel formed is highly viscous. As the fibres aggregate, they align to form bundles which increase their rigidity as well (Figure 1.15).

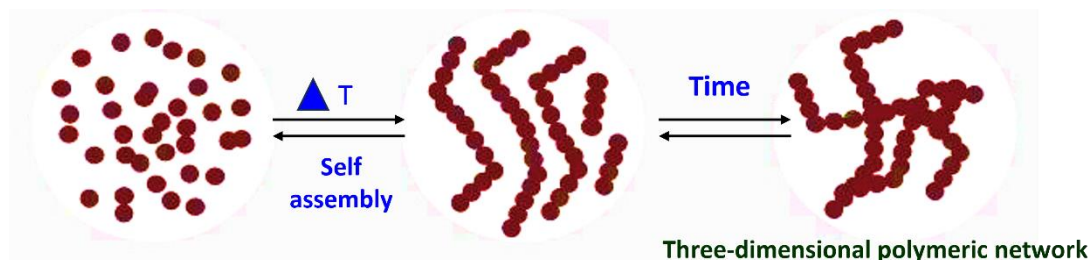


Figure 1.15 Illustrative diagram of organogel formation through solid matrix mechanism

Based on the growth of fibres, the solid aggregates form either unidimensional or bidimensional. Aspect ratio with higher length to lower width experiences aggregation via one dimensional growth. They are usually in the shapes of rods and tubes ranging 10 μm -100 μm ratio. These branched one-dimensional network structure further assembles to form SAFiN (self-assembled fibrillar network) [101]. While for two-dimensional growth, the aggregates form micro platelets. The structural form of these aggregates is crystalline while following heterogeneous or homogeneous arrangements. The solid fiber mechanism is widely used for most of the LMOGs as strong gels are formed from permanent and crystalline networks.

1.3.3 Polymeric matrix

High molecular weight gelators display a high capability of gelling diverse sets of organic solvents even at minimal concentrations. The polymer chain crosslinks and interacts through covalent bonding while undergoing ionic interaction in the presence of a polymeric gelator to form three-dimensional network structure [45]. However, the nature of the crosslinking may either take place physically or chemically. The presence of functional groups in the chain connects to crosslink forming three-dimensional network through covalent bonding. Due to the polymeric network formed between gelator particles and solvent molecules, they can trap organic solvents within the network frame. Also, the involvement of solvent obstructs the structure from collapsing. Due to the nature of crosslinking, the organogel renders a stable nature (Figure 1.16).

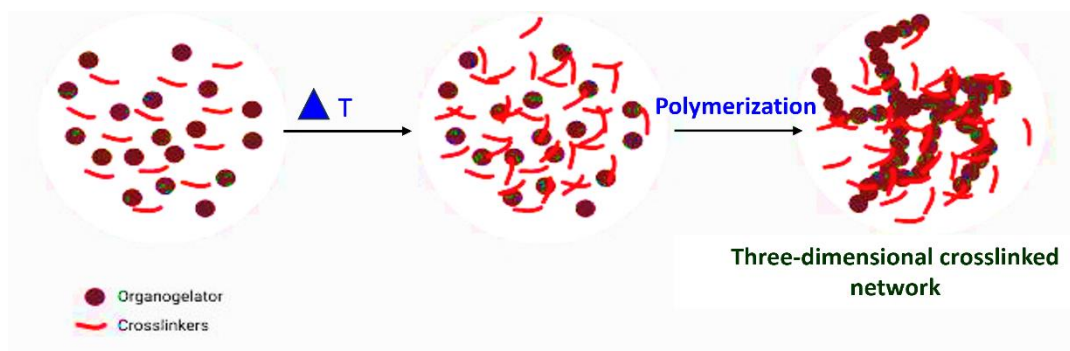


Figure 1.16 Illustrative diagram of organogel formation through polymeric matrix mechanism

While organogels prepared using copolymers of methacrylic acid, diacetylene, methyl methacrylate, ethylene are some of the commonly used polymeric gelators. The formation of supramolecular polymeric organogel can also be observed as gelation was induced due to the interaction of LMOG with the polymeric gelator [84]. The presence of LMOG initiates the self-assembling of the polymer molecules while the polymeric backbone is involved in the gelation procedure providing substantial space for obstructing the flow of solvents. However, crosslinking in these organogels may occur due to non-covalent bonding and may display dynamic reversibility [102].

1.3.4 Emulsion matrix

Dispersion of two or three liquid droplets which are immiscible in nature forms emulsions. Formed in two phase system, a simple emulsion of oil in water (o/w) or water in oil (w/o) modifies to form a heterogeneous system of multiple emulsions (w/o/w or o/w/o) (Figure 1.17) [103]. The presence of dual surfactant lipophilic and hydrophilic stabilizes the system using electrostatic, structural-mechanical, and fluidic (Gibbs-Marangoni) interaction [104].

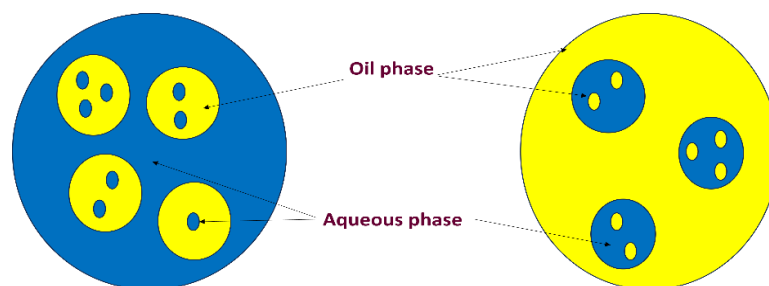


Figure 1.17 Illustrative diagram of organogel formation through emulsion mechanism

Rearrangement of surfactants at the droplet interface exposes hydrophobic and hydrophilic moieties to their respective oil and water phases. The ability of hydrophilic drug to get enclosed within the water phase of a w/o/w emulsion makes the w/o/w highly suitable and widely used mechanism for delivery of bioactive ingredients [104]. Development of multiple emulsions follows phase inversion (one step technique), double emulsification technique (two step emulsification), membrane emulsification and microchannel emulsification techniques [105]. However, among the four, double emulsification is highly preferred due to its stability and greater encapsulation efficiency. This renders them highly potential for use in controlled drug release, separation science as well as enzyme mobilization.

1.4 ORGANOGEL FOR WATER PURIFICATION

Organogels, composed of organic, non-polar solvents, demonstrate remarkable thermal stability and insensitivity to moisture, rendering them resistant to microbial contamination [106]. These properties, in turn, make them ideally suited for environmental applications, particularly in water treatment.



Figure 1.18 Water contamination caused by release of (a) dye molecules from textile industries (<https://textilelearner.net/effluents-from-textile-industry-and-it-impacts-on-environment>), (b) oil spillage in river banks (<https://education.nationalgeographic.org/resource/point-source-and-nonpoint-sources-pollution>)

Additionally, organogels exhibit the ability to absorb solvents through swelling, as well as undergo surface modifications for adsorption on the gel surface. As such, organogels represent a promising platform for a range of applications, from environmental remediation to tailored adsorption.

As technology and industry continue to evolve, the contamination of water resources has emerged as a pressing concern in neighboring areas. The textile and paper industries specifically rely heavily on dyes to impart color, but more than 80% of their effluents are discharged onto water surfaces, polluting the environment with toxic extracts that can cause carcinogenic issues (Figure 1.18). These dyes are highly soluble in water and challenging to separate from it. Their structure, which includes multiple aromatic rings, makes them susceptible to biodegradability content. Certain implications of organogels in water treatment are as discussed below:

1.4.1 Organogel for dye adsorption

A crosslinked polymeric organogel, dimethyl 2-(methacrylamido) pentanedioate, was synthesized by crosslinking glutamic acid with diethylene glycol dimethacrylate. The organogel did not swell in water and was used to adsorb hydrophilic dyes. Additionally, the organogel showed solvent uptake behavior in solvents such as DMF, THF, acetone, DCM, and methanol due to its porous network. Figure 1.19(i) illustrates the removal of dyes CV, RhB, and MG before and after adsorption using the solvent [107]. Another organogel was prepared by chemically crosslinking a deblock copolymer. The structure of this organogel consists of interconnected spherical micelles organized in a random percolated network structure. The presence of N^+ ions in the micelle structure enables the organogel to act as an adsorbent for anionic dyes, demonstrating anionic selectivity in a mixture of cationic and anionic dyes. Over 90% of the dyes were removed within 24 hours, and the organogel exhibited excellent reusability [108]. Figure 1.19(ii) displays the adsorption of dyes MO, CR, brilliant blue R-250, and bromocresol green by the adsorbent.

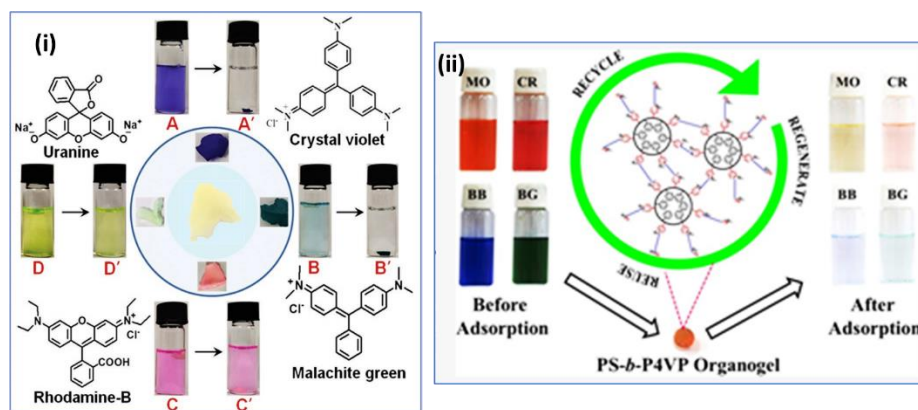


Figure 1.19 Application of organogel in the removal of dyes, before and after adsorption [107,108]

The recent advancement in the field of peptide-based gelators as intelligent soft materials has paved the way for their application in addressing environmental concerns. As an illustration, a peptide organogel based on tryptophan has been employed to effectively eliminate both cationic and anionic dyes, as well as toxic metals [109]. Utilizing p-aminobenzoic acid as a peptidic gelator has yielded mechanically robust thermoreversible organogels. Due to its electron-rich properties, this gel demonstrates remarkable efficiency in adsorbing cationic dyes from wastewater, achieving a removal efficiency of more than 90% [110]. Furthermore, the self-assembly of tripodal pyridine has enabled the formation of weak non-covalent gels, which can be modified to generate stable covalent supramolecular gels. These gels possess positively charged surfaces and a porous structure, rendering them excellent adsorbents with a high capability to adsorb anionic dyes [111].

Another effective adsorbent for the elimination of both cationic and anionic dyes is a polymeric organogel prepared through the reaction of cellulose diacetate, derived from plants, with PVA. The presence of a porous structure facilitates host-guest interactions, resulting in enhanced dye adsorption [112]. PVA acts as the polymer framework and provides hydroxyl groups for the formation of reversible boronate ester bonds. This self-healing smart organogel, synthesized via dynamic covalent bonding, exhibits the ability to adsorb dyes such as MB, RhB, and MO. Under basic conditions, boric acid becomes negatively charged, enabling it to interact with the positively charged centers of the dyes [113].

1.4.2 Organogel used for metal adsorption

Recently, there has been a growing interest in the potential of organogels to remove heavy metal ions. Roy et al. conducted a study in which they successfully formed an organogel using gemini and hetero-gemini gelator molecules based on nicotinic acid. The presence of functional groups in the structure enabled the organogel to detect Cd(II) and Pb(II) ion [114]. In another study, the combination of dual gelator molecules was found to enhance gel strength and improve stability in aqueous solutions. Consequently, the organogel was capable of removing Hg(II), Cd(II), and Pb(II) by forming metallogels with their respective salts [115]. Lin and colleagues reported the formation of a supramolecular organogel called BTG, which exhibited strong aggregation-induced emission. This organogel demonstrated exceptional adsorption of Cu²⁺ ions, with a removal efficiency of 99.02% [116].

Sureshan and group reported the formation of an organogel using crown ether (CE) and diisopropylidene mannitol (SG6). The CE gelator assembled through weak non-covalent interactions (NCI), while SG6 assembled more rapidly due to stronger hydrogen bonding, resulting in the formation of fibrils. These fibrils attracted CE to the surface through hydrogen bonding, facilitating their self-assembly via weak NCI. When styrene-based polymeric gels were formed using the combination of the aforementioned gelators, washing led to the formation of robust porous hybrid PS-CE hybrid matrix networks, with SG6 leaving behind pores. The presence of CE in the formed organogel was able to selectively bind alkali metals as Li⁺, Na⁺, K⁺ [117].

1.4.3 Organogels for solvent absorption

Organogels have the unique ability to swell in organic solvents, allowing them to form a three-dimensional network. Due to their high uptake capacity, this network can effectively function as a sorbent. In particular, the network is able to selectively absorb non-polar solvents while holding other solvents apart. The three-dimensional network formed by organogels thus exhibits significant potential as a sorbent material. If gel is relatively high, the three dimensional network are able to hold solvents apart from selective absorption for non-polar solvents [37]. Bayraktaroglu and group demonstrated the formation of hydrophobic sorbent prepared from PDMS and tetraethylorthosilicate. The sorbent showed excellent absorption in most of the organic solvents with the highest

being 2119% in DCM (Figure 1.20i). The sorbent was able to separate mixture of solvents and water as the water affinity was comparatively low [118].

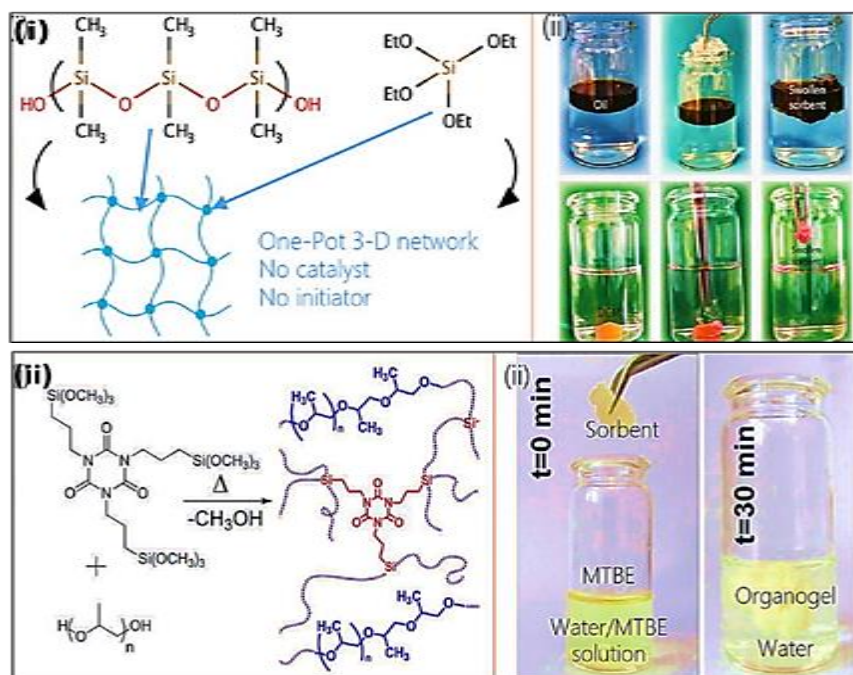


Figure 1.20 Application of solvent absorption by organogels: (i) Structure of the formed polydimethylsiloxane sorbent and their removal of DCM and crude oil[118] (ii) Structure of the formed PPG-ICS organogel and absorption of MTBE from aqueous solution[119][37]

A covalently bonded network of organogels could selectively adsorb aromatic components apart from separating aromatic solvents from azeotropic mixture [120]. Goel and Jacob reported the formation of organogel using acrylate based linear and crosslinked polymer with D-galactose capable of absorbing non-polar solvents with highest swelling of chloroform by 935% [121]. Demirel et al. performed synthesis by varying the molecular weight of polypropylene glycol (PPG) and tris [3-(trimethoxysilyl)propyl] isocyanurate. As the weight of PPGs increased, a decrease in crosslinking was observed followed by increase in absorption capability. DCM observed highest absorbance of 1872% while gasoline and MTBE absorbed 770 and 714% respectively (Figure 1.20ii) [119]. Kizil and Sonmez reported the synthesis of amphiphilic crosslinked polymeric network of sorbent. The sorbent was prepared using a branched PEG and monomers of organosilane through bulk polymerization. Owing to presence of amphiphilic structure, the sorbent posed as organogel can intake organic solvents as DCM with 1829% [122]. Similar work was prepared through interaction of a cyclic aliphatic glycol and aromatic alkoxy silane group. The sorbent swelled around 641% and 747% in toluene, and THF respectively [123].

The utilization of hydrophilic monomers has been reported to induce extraordinary swelling behavior in hydrogels [124]. In this regard, we have employed similar hydrophilic monomers to investigate swelling properties in prepared organogels. The use of PANi gels has witnessed a rapid rise due to their wide applicability in electric devices, owing to their conducting properties. With reference to the synthesis of elastic PANi/PVA composite-based organogel reported by Wang and his group [125], we also sought to incorporate PANi to study swelling and other applications, inspired by the aforementioned works.

1.5 OTHER APPLICATIONS OF ORGANOGEL

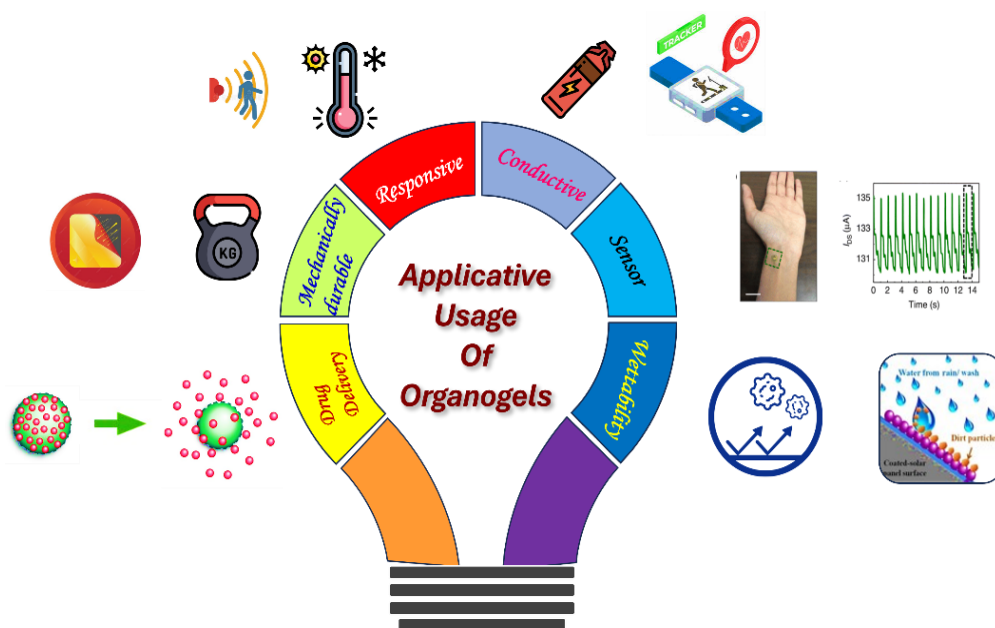


Figure 1.21 Schematic illustrations of applications related to organogels

Based on their physicochemical properties, organogels have been implemented over a wide range of applications. Since more stable and robust organogel with high mechanical strength are observed due to formation of covalently bonded network in chemical organogels, they are utilized for industrial applications as anti-icing, anti-fouling, adhesives, highly durable gels [32]. While physical organogels observe weaker intermolecular interaction yielding lower mechanical strength as well as stability. These gels are thus used in applications involving food processing, drug delivery, cosmetics, bioelectronics, pharmaceuticals (Figure 1.21). Following characteristics attributes the applicative usages of organogels.

1.5.1 Organogels in drug delivery

The inadequate solubility of numerous drugs in aqueous phase allows the usage of organogel for entrapping bioactive compounds suitable for transdermal [126], oral [127], intravenous [128] routes for drug delivery. Lu and group prepared a nanoemulsion-based organogel loaded with capsaicin. Prepared organogel witnessed the treatment of capsaicin intake through oral route by increasing bioaccessibility and improving the rate of dissolution [129]. Dai et al. prepared an organogel loaded with flunarizine hydrochloride through an insitu process for delivery of intraocular drug while treating brain afflictions [130].

1.5.2 Mechanically durable organogels

Due to confinement of organic solvents within the three-dimensional structure, the swollen gels gets fragile and are prone to brittle nature due to decreased interaction between the formed chain of network [69]. Park and co reported the introduction of aromatic compounds to the network structure of the organogel. The aromatic interaction observed a rise in swelling ratio while enhancing plasticization of the formed network with twisting, folding, bending and stretching in comparison to non-aromatic interaction [131]. A mechanically durable, tough gel was prepared by Mo et al. as the formation of dual polymer network occurs due to the formation of H-bonding between ethylene glycol and hydroxyl groups of polyvinyl alcohol and polyacrylic acid [132]. Excellent adhesive property was observed by the organogel as the gel was able to stick to surfaces like glass, rubber and metal surfaces supported by complex formation due to presence of borate bonds.

1.5.3 Responsive organogels

Due to the presence of organic solvents and formation of three-dimensional network, organogel responds to stimuli and flexibility including changes in volume and shape [133]. This makes the organogel behave as actuators where the inclusion of responsive fillers corresponds to quick and effective actuation. Fabrication of a liquid crystalline based organogel was reported by Jin and group. On controlling the phase evaporation, the morphed structure of solvent infused organogel can be reprogrammed, post programmed or even erased depending upon the gel alignment [134]. Owing to the transition involving temporary and permanent formation of shapes, such organogel are found to be stable during actuation, leading to switching shapes reversibly from kirigami and origami upon treatment with cooling and heating respectively.

1.5.4 Conductive Organogels

Zhang and his group attained similar properties in an electronic skin sensor-based organogel suitable for biomedical devices. Silver nanowires capped onto PVP were physically and chemically crosslinked using polyacrylamide forming double networked ethylene glycol-based organogels [135]. This e-skin organogel experienced temperature tolerance from -20 to 80 °C with excellent tensile strain and exhibiting stretchability of > 22000%. Owing to their quick responses and superlative electrochemical stability, they are preferably used as electrolytes in batteries or as stress sensors presenting promising applications in wearable electronics.

1.5.5 Organogels as sensors

As the polarities vary widely in organic solvents contrasting to aqueous phase of hydrogels; organic molecules suitable for sensing or detection purpose can be stuffed onto organogels for enhancing performances. An ionic organogel based on polyacrylamide was prepared by Liu et al. for observing interactive color change [136]. With the inclusion of an electrochromic molecule within the polymer network, the gel formed was able to trace stimuli in human body through change in color, making it effective as wearable device for detecting motion in finger flexion or wrist. Smith and group developed a protein-based stimuli responsive organogel by subsequently changing the solvent followed by binding of ligands to the protein molecules [137]. The protein based organogel observes volume phase transition which on binding with ligand molecule influences macroscopic transitions thus swelling up the organogel. These protein binded ligands acts as sensors for quantitating binding of ligand in drug samples.

1.5.6 Organogel exhibiting wettability

Since organogels are solvent induced materials, surface of the gel evidences properties as anti-icing, anti-fouling, droplet manipulation, self-cleaning. Wang and co developed a thermoresponsive droplet adhesion system [138]. With the infusion of lubricant on the organogel surface, the hydroxyl group present in single-stranded DNA interacts with the lubricants controlling the sliding property. While with the increase in temperature, hydrophobic interaction could significantly reduce leading to smoother sliding on the surface of the organogel. An amphiphilic organogel with anti-icing property was prepared by Yu and group [139]. Developed using poly(PDMS-r-PEG-r-GMA) copolymer and DADPDS, the infused amphiphilic lubricant provided anti frosting

and anti-icing ability due to the synergistic effects caused by the lubricant. Due to the hydrophobic chains of PDMS, the organogel remained stable under heat as well as underwater. While the hydrophilic segment of the amphiphilic lubricant formed H-bonding with water molecules hindering ice formation on surface. Thus, providing durability and stability to the gel to be used for practical applications.

1.6 SCOPE AND OBJECTIVES

The physicochemical properties of semi-solid materials can be significantly enhanced through the use of organogels, which are influenced by their structural and chemical composition. The type of network formed and the solvent of choice play a key role in achieving this desirable outcome. As such, careful consideration of these factors is necessary in the design and formulation of organogels, particularly in the context of their potential application in various industries [32].

With the growing increase in industrial sites, disposal of waste products involving organic and inorganic pollutants onto water bodies have left the aquatic sources in a deplorable site. Industrial waste also comprises organic solvents and inorganic contaminants such as heavy metals and hydrocarbons. Eliminating these toxic pollutants is a strenuous activity, as they can persist for a prolonged period, adversely impacting both aquatic and terrestrial life and polluting our environment. Exploring the unbounded potential application of gels from pharmaceutical breakthroughs to the industrial sector imparts the importance of gels as irrefutable in the era of modern science. To separate the pollutants from water sources, organogel as an effective material has been adopted for remediation purpose. Although implemented for practical applications involving sensors, conductivity, durability followed by usage in cosmetics, drug delivery as well as pharmaceuticals; very few works have been reported concerning environmental issues. Due to the ease of preparation while the formation of moisture-insensitive and thermally stable gels, organogels can be implemented for absorption along with the adsorption of organic and inorganic pollutants. Utilizing these network structures of gel, works involved in this thesis endeavor on the formation of organogel, their synthesis, characterization, and investigation of the efficacy of organogel as a removal mechanism for such pollutants, through the use of both adsorption and absorption methods.

Motivated by the ongoing discussions, the following objectives are set for carrying out the research work.

Objectives of the present investigation:

- ❖ To synthesize
 - (i) Polyvinyl alcohol grafted Stearic acid (PVSA) organogel
 - (ii) Polyvinyl Alcohol crosslinked Acrylamido methylpropane sulphonic Acid grafted Stearic Acid (PACSG) organogel
 - (iii) Polyvinyl alcohol-modified Aluminum Montmorillonite (clay) / Polyaniline (PMCP) hybrid composite-based organogel
- ❖ To study the morphological, thermal, and mechanical properties of the synthesized organogel
- ❖ To characterize the organogel using analytical techniques FTIR spectroscopy, scanning electron microscopy (SEM), X-ray diffraction (XRD and HRXRD), thermal analysis (TGA, DSC), spectroscopy (UV-Vis)
- ❖ To explore applications of organogel for water purification
 - (i) Application of the organogel in swelling analysis of different aromatic and hydrocarbon solvents
 - (ii) Removal of cationic and anionic dyes from contaminated water sources using organogel
 - (iii) Removal of metal from water using organogel through adsorption.

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