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Exploration of Organogels and their Applications in Materials and Biology

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Abstract

Organogelators belong to an essential class of soft matter which has received increasing attention in recent years due to their easy fabrication into soft materials having utility in both industrial and pharmaceutical applications. Sugars and amino acids being highly hydrophilic, assemble poorly with organic solvents. However, with suitable derivatization with aromatic rings, long alkyl chains, and other hydrophobic moieties, they can be built into environmentally benign excellent organogelators with inherent chirality, complex structural miscellany, and biocompatibility. This review contains a refined literature collection corresponding to the recent advancements in the design and synthesis of a number of important carbohydrate and amino acid-based organogels along with their potential applications in various industrial and biomedical sectors, such as photo-and chemo-sensing, organic, and optoelectronics, cosmetics, drug delivery, and cancer treatment, etc. (Figure 1)

Keywords: Organogels, gelators; self-assembly; weak interactions; carbohydrates; amino acids; low molecular weight organogelators; drug delivery; sensors

Abbreviations: LMOGs: Low Molecular Weight Organogelators; LMHGs: Low Molecular Weight Gelators; PEG: Polyethylene Glycol; MST: Monosaccharide Transporter; CGC: Critical Gel Concentration; DMAP: Dimethyl Aminopyridine; PSOG: Phase Selective Organogelator; RAFT: Reversible Addition-Fragmentation Chain Transfer; DEGDMA: Di(Ethylene Glycol) Dimethacrylate; HEMA: 2-Hydroxyethyl Methacrylate; PG: Propylene Glycol; DEPC: Diethyl phosphoryl Cyanide; MGCs: Minimum Gelation Concentrations; FAs: Fatty Acids; Phe: Phenylalanine; NMP: N-Methyl Pyrrolidone; DMSO: Dimethyl Sulfoxide; OLED: Organic Light Emitting Devices; Hepg2 Cells: Hepatocellular Carcinoma Cells; FNZ: Flunarizine Hydrochloride; CTM: Clotrimazole; ACV: Acyclovir; GF: Glycerol Formal; NSAIDS: Non-Steroidal Anti-Inflammatory Drugs; Plos: Pluronic Lecithin Organogel; SMP: Sorbitan Monopalmitate; SFA: Saturated Fatty Acids; TFA: Trans Fatty Acids; Chn: Chitin Nanocrystals; PDBA: 1,4-Phenylenediboronic Acid; FE-SEM: Field Emission Scanning Electron Microscopy; DEPC: Diethylphosphoryl Cyanide

Introduction

The term "gel" is derived from the word "gelatin." It is believed that the words "gel" and "jelly" both descended from the Latin word "gelu," which means frost, freeze, or solidify. Thus, "gel" originates from a liquid condensing into a solid-like or semisolid substance that cannot flow. Due to their elastic or semi-elastic nature, these materials maintain some features of liquids [1]. Gels are soft semi-solid substances made up of many materials, especially those that serve as a liquid dispersion medium (referred to as the "solvent") and the gelling agent (gelator), with the former usually constituting a more considerable numerical portion of the whole [2]. Gels are the interface between complicated fluids and phase-separated states of matter, a class of soft matter systems. At room temperature, the gelators were insoluble in almost all solvents. However, they dissolve upon heating and get gelled upon cooling. By measuring the minimum concentration of gelator necessary for the formation of a stable gel at 25°C, the critical gelation concentration (CGC) was ascertained [3]. Many present and ancient human technologies include gels as an essential component. Since the Middle Ages, natural pectin, gelatin, and agar gels have been used in food products. Gels can also be found in lubricants, explosives, soap, adhesives, cosmetics, and medical implants. Though several definitions are known in the literature, the universally accepted IUPAC definition is as follows "A colloidal network that is enlarged throughout its entire volume by a fluid".

Water, organic solvents, or ionic liquids can all be used to create gels, referred to as hydrogels, organogels, or ion gels. However, aerogels, on the other hand, are gels that occur when their fluid state is a gas [4].

The self-assembly of gelator molecules confines them proximally and spatially, generating microstructures, such as fibres, vesicles, and so on, preventing the fluidity of the solvent from resulting in gelation [5]. Because they have intriguing physical characteristics like transparency, thermos responsiveness, viscoelasticity, and moldability, gels, 3D soft materials that display qualities halfway between those of liquids and solids, are essential academically and industrially. In particular, gels can be divided into two main categories: physical gels and chemical gels. Physical gels are created when low-molecular-weight organic molecules self-assemble in suitable solvents through weak intermolecular interactions, such as hydrogen bonds, electrostatic interactions, van der Waals interactions, pi-pi interactions, etc. They are further classified into two main categories: organogels, which are congealed using organic solvents, and hydrogels, often known as aquagels or water-based gels [6]. The general classification of gels has been given in Figure 2.

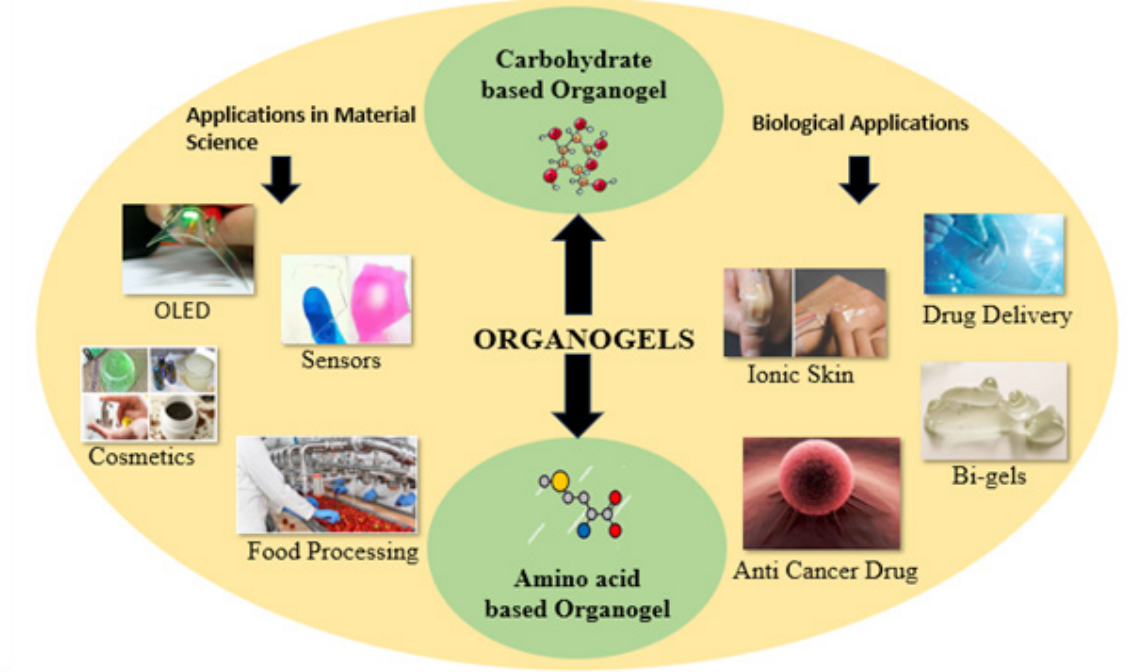

Figure 1: Graphical abstract.

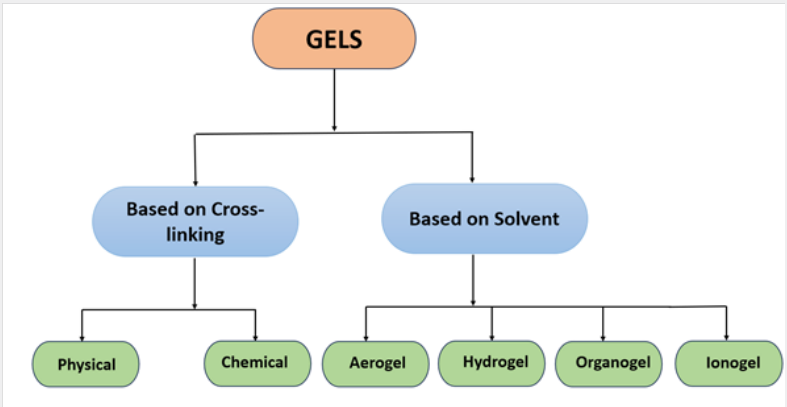

Figure 2: Classification of gels.

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Hydrogels

Hydrophilic crosslinked three-dimensional networks of hydrogels can collect and hold significant amounts of water molecules through hydrogen bonds before releasing them reversibly. The hydrogel frameworks are reactive and can exchange components and data with the surroundings. Hydrogels can react to various external stimuli, including pH, temperature, light, electric field, magnetic field, biomolecules, etc. The soft and wet characteristics are comparable to biological tissues [7]. Various materials, including synthetic polymers like polyvinyl alcohol and polyacrylic acid, natural products like alginates and proteins, copolymers, or wild and synthetic polymers, can be exploited to produce hydrogels [8]. In the case of polymeric compounds, the hydrogels' mechanical and chemical properties will be significantly influenced by their monomers, crosslinkers, water content, and crosslinking densities. Due to the complex composition and insolubility of the cross-linked hydrogels, it is challenging to characterize structures like molecular weight precisely. Nevertheless, due to their homogeneous and isotropic systems, conventional hydrogels often need better mechanical properties and react slowly to stimuli, significantly restricting their uses. Several methods have been established using secondary bonds, topological slide-ring structure, nanocomposite reinforcement, and supramolecular interactions to increase toughness and response rate [9]. The biomedical industry is highly interested in hydrogels based on natural polymers of polysaccharides and protein, especially for tissue regeneration and drug delivery [10].

Organogels

Organogels are produced when surfactant molecules (organogelators) self-assemble into three-dimensional networks that enclose an organic liquid using capillary forces. Based on the type of gelator applied: polymeric or low molecular weight organogelators (LMOGs), they can be formed by physical forces and chemical interactions. Since the ability to make supramolecular gels is only attainable with particular materials, gelator structure significantly impacts organogel characteristics [11]. Low molecular weight gels are a gel category created by non-covalent interactions. They are also known as physical gels or supramolecular gels. Low molecular weight gelators can be divided into low molecular weight hydrogelators (LMHGs) if the solvent is water and low molecular weight organogelators (LMOGs) if the solvent is an organic solvent or a mixture of an organic solvent and a solvent that is aqueous [12]. LMOGs can self-assemble to generate organogels using organic solvents [13].

Various molecular ionic liquid gels have been formed using affordable, sustainable, and easily synthesized sugar-based low molecular weight gelators from isosorbide and mannitol. Electrochemical applications are particularly interested in this class of reversible gels. Low molecular weight gelators generated from sugars have been demonstrated to gel a wide range of solvents from water to oils; this may be owing to the development of strong hydrogen bonds, as such interactions have been recognized as a significant factor in gel formation. In addition, sugars give rise to homochirality, which is often essential for gel formation [14]. Due to their multiple potential uses in stimuli-responsive materials, intelligent electronics, oil-spill recovery, medication delivery, and other fields, low molecular weight organogels have received a lot of research interest during the past several years. LMOGs self-assemble into fibrous structures, tapes, sheets, etc., because of intermolecular hydrogen bonding, -stacking of aromatic units, hydrophobic effect, and van der Waals forces. The suspension of the solvent, often known as a gel, is the end outcome of connecting these secondary structures. Amino acids offer a wide range of applications such as LMOG because they are adaptable compounds for self-assembled structures [15].

Organogels are an acceptable replacement for high-saturated and trans-fat food choices. Organogels, and lipid gels, have multiple uses inside and outside the food industry. According to research, these organogels may limit oil migration in chocolate and govern the release of health-beneficial reactive substances such as antioxidants and bioactive compounds. Organogels have been found to improve oil oxidative stability by enclosing the oil in a gel network, resulting in less exposure to oxygen. Specific celluloses, waxes, and phytosterols, such as a β-sitosterol/γ-oryzanol combination, are effective organogelators for food applications [16]. Anthracene, anthraquinone, and steroid-based molecules are examples of organogelators that cannot form hydrogen bonds. At the same time, amino acids, amide, urea moieties, and carbohydrates are examples of organogelators capable of forming hydrogen bonds [17]. The most popular initial organic solvents designed to prepare organogels were alkanes with more than five carbons, such as hexane, cyclohexane, and alkene squalene. Cosolvents for organogels can also be polar solvents like water, ethanol, or polyethylene glycol (PEG). The most popular solvents utilized in the production of organogels are glycerol, PEG 400, ethylene glycol, and propylene glycol. Organogels are frequently made with heat, but before they are combined with hydrogels, the mixture of their constituent parts is typically cooled to room temperature. Hydrogels produced at high temperatures may occasionally undergo this cooling process [18]. Compared to hydrogels, organogels are fascinating because of the highly customizable characteristics brought on by the inherent benefits of diversity in the gel components. The development of accurately altering the solvent part in organogels has been substantially enhanced by modifying compound solvent systems, such as nonpolar-polar organic liquid mixture systems and polybasic organic liquid systems [19]. (Figure 3)

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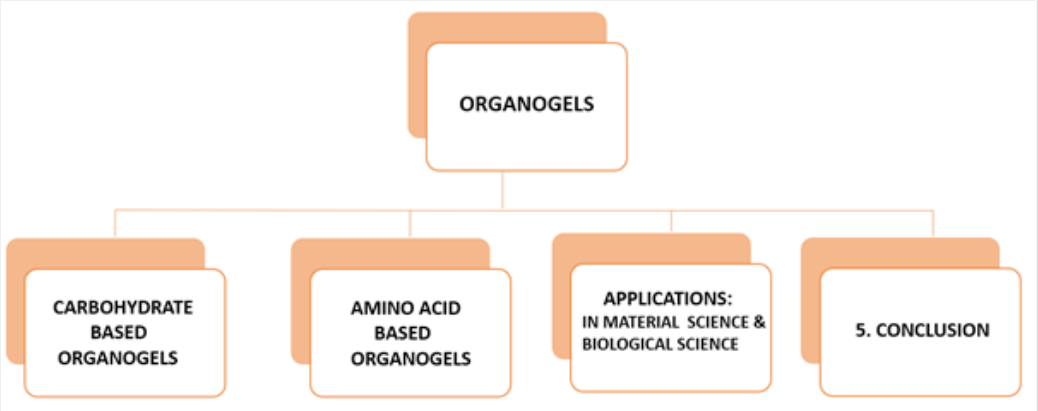

Figure 3: Overview of the article.

Carbohydrate-Based Organogels

Carbohydrates are the most common type of biopolymer, primarily employed for energy production and as structural materials [20]. Of these, monosaccharides are the most basic units for forming complex macromolecules, and glucose is undoubtedly one of the most essential carbohydrates for life [21]. These complex biomolecules may create many hydrogen bonds, making them appealing for supramolecular systems [20]. Carbohydrates are adaptable self-assembly building blocks because they range from basic sugar units to sophisticated granular starch-structure-like organization of (1,4)-a-linked glucans [22]. Carbohydrates have a general empirical structure of $(CH_2O)_n$ and are simple monosaccharides in their basic form. These essential elements can unite to generate more complex sugars. A disaccharide is created when two monosaccharides are joined. Carbohydrates of two to ten simple sugars are oligosaccharides, whereas longer chains are polysaccharides [21]. Carbohydrates are hydrophilic building blocks that can establish multiple H-bonds due to hydroxyl groups' presence, promoting water solubility. Because of their directed hydroxyl groups, cyclic forms of carbohydrates produce more stable gels, allowing cooperative networks to self-assemble into fibrous structures [23]. Some of the essential carbohydrates present in natural sources are shown in Figure 4.

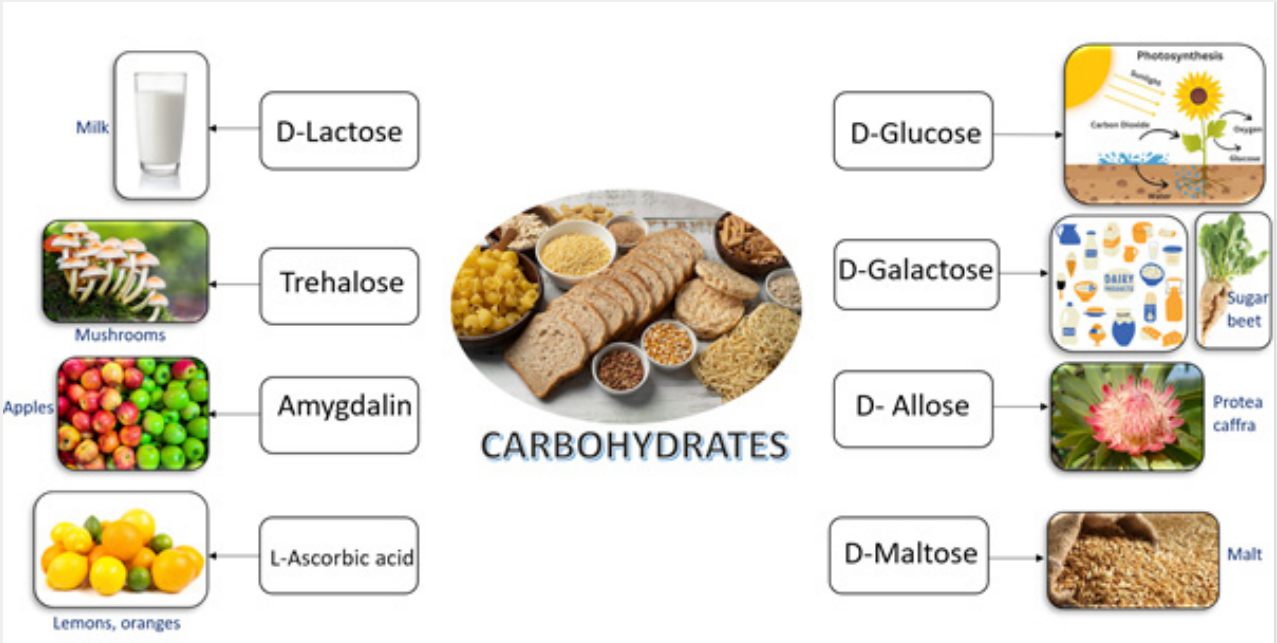

Figure 4: Natural sources of some essential carbohydrates.

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Many carbohydrates have been used as organogelators because partially protecting carbohydrates as cyclic ketal derivatives impart amphiphilicity and aids in preorganization by constraining the conformational freedom for self-assembly [24]. Supramolecular gels generated by carbohydrate-derived low molecular-weight gelators are intriguing soft materials with many applications [25]. Carbohydrates are frequently employed in synthesizing low molecular weight gelators because they are abundant and contain many chiral centres that can be selectively functionalized. In addition, because of their commercial availability, low cost, strong biocompatibility, and low environmental impact, they have emerged as an attractive feedstock for the design and synthesis of low molecular weight gelators. Several effective gelators have been developed and produced using glucose and glucosamine as building blocks [25,26]. Low molecular weight carbohydrate gelators are an intriguing family of compounds with numerous applications. Sugar-based organogelators are valuable for drug administration, enzyme immobilization, and other applications [27].

In a single step, three new sugar-based low-molecular-weight gelators, shown in Figure 5, were created by combining glucose units with hydrocarbon chains. These artificial glucose derivatives act as gelators for a range of solvents, including water. These low molecular weight gelators made from glucose are thermotropic and thixotropic by nature, and this thixotropic behaviour was seen in the organogels derived from these low molecular weight gelators right after the functionalization with 4,6-O-benzylidene at moderate loadings [28].

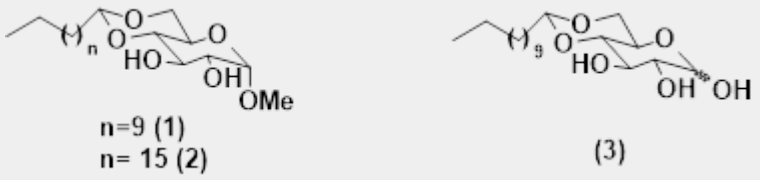

Figure 5: Molecular structures of glucose derivatives (1-3) [28].

Because of the simple synthesis and the amide-like characteristics of the triazole heterocycle, 1,2,3-triazole-containing molecules are beautiful among carbohydrate derivatives. A variety of carbohydrate and triazole derivatives have been synthesized using the copper (I) catalyzed azide-alkyne cycloaddition reactions (CuAAC), the "click chemistry," and they have shown a wide range of applications in synthetic chemistry, medicinal chemistry, biochemistry, and catalysis [29-31]. Several monosaccharides and disaccharide-based triazole compounds synthesized by click chemistry have recently been demonstrated to be effectual low molecular weight gelators. Per-O-acetylated D-glucose and D-glucosamine derivatives were potent low molecular-weight gelators. Other applications for sugar-based triazole-derived organogelators include oil spill cleanup and dye removal [32]. Most carbohydrate-based low molecular weight gelators are structurally based on glucose, galactose, and glucosamine; however, a few examples of organogelators based on mannose and xylose have also been observed [33]. A significant number of organogelators are synthesized from carbohydrates, particularly monosaccharides and polysaccharides.

Monosaccharide-Based Organogelators

The natural abundance of sugar derivatives emphasizes their importance in nature. For example, one of the significant monosaccharides, D-glucose, supplies energy to the cells [34]. Because it is relatively straightforward to create substituted products by selectively functionalizing the anomeric position and the 4- and 6-hydroxy groups, glucose is a versatile building block for synthesizing diverse small molecule gelators [25]. Sugars are essential to all life as sources of nutrition and signalling chemicals. Plants rely on controlled sugar intake for proper growth of organs and sugar preservation, and apoplastic sugar depletion is a defensive tactic against microbial diseases such as rusting and fungus. Sugar Transport Proteins, proton-coupled symporters from the Monosaccharide Transporter (MST) superfamily, enable glucose and other monosaccharide transport. Photosynthetically produced sugar in plants is mainly transported as sucrose throughout the plant body via the phloem. The conversion of sucrose by invertases to glucose and fructose, and then its transmembrane uptake into sink cells controlled by Sugar Transport Proteins, is an effective two-step mechanism in apoplastic sugar import from the phloem [35]. Small molecule gelators with a carbohydrate basis are significant families of compounds that can create valuable soft materials with vaat utilities in nature. The molecular self-assembly of various derivatives of common monosaccharides, such as D-glucose and D-glucosamine, has provided insights into the structural requirements for successful molecular gelators (4-6), as shown in Figure 6 [36].

One essential monosaccharide preservation method is synthesizing their 4,6-O-benzylidene derivatives, which can be building blocks in expanding sugar chains [34]. Certain 4,6-O-benzylidene acetal-protected monosaccharide derivatives are potent sugar-based LMWGs. Products of 4,6-O-benzylidene-

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methyl-α-D-glucopyranose, the derivatives (7-11) given in Figure 7, for example, had good gelation characteristics [27,36]. Several investigations have concentrated on the gelation capabilities of di-O-benzylidene-monosaccharides as one of the earliest carbohydrate-based organogelators [37]. Carbohydrates are also nature's most abundant renewable resource. Wang and several other researchers have documented multiple classes of efficient carbohydrate-based low molecular weight organogelators and hydro gelators, taking advantage of the abundance of natural monosaccharides and disaccharides. The structures (12-16) given in Figure 8 are Common sugars such as D-glucose, D-glucosamine, N-acetyl-D-glucosamine, D-lactose, D-Galactose, D-Mannose and D-maltose, including sorbitol, belong to them [39].

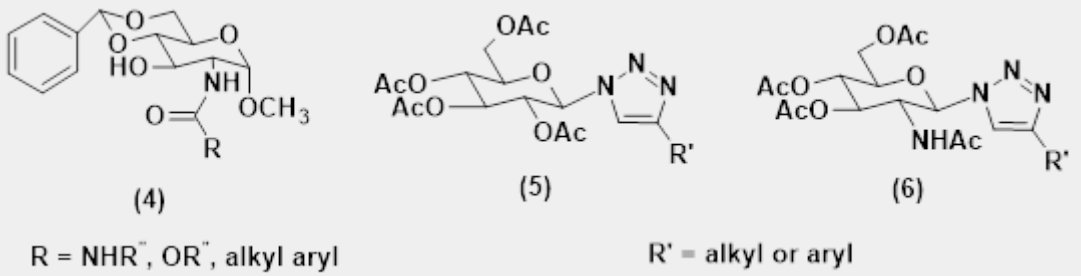

Figure 6: Representative examples of D-glucose and D-glucosamine gelators (4-6) [36].

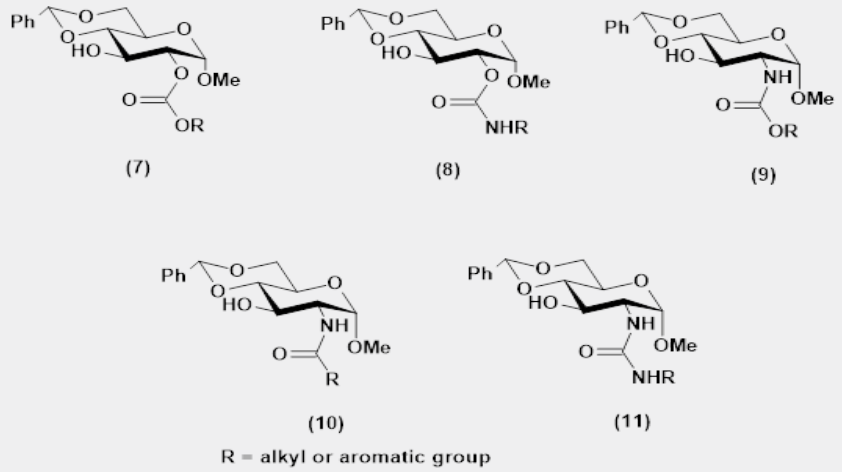

Figure 7: Derivatives of 4,6-O-benzylidene-methyl-α-D-glucopyranose structures altered at the second position [38].

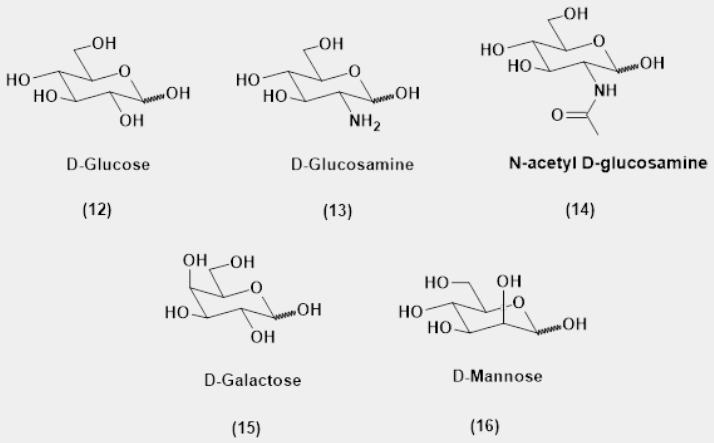

Figure 8: Sugar starting materials structures (12-16) are often used for scheming low molecular weight gelators [39].

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Mannose-Derived Carbohydrate Gelator

K.B. Pal and coworkers had established a simple mannose-derived carbohydrate gelator (Figure 9) with various functional features, and this thermo-reversible organogelator has exceptional self-healing ability. At room temperature, this gelator exhibited phase-selective gelation behaviour and formed organogels from biphasic oil-water or organic liquid-water combinations [40]. The well-known p-methoxyphenyl mannose (17) has been processed with 2,2-dimethoxy propane in dry acetone in the presence of H_2SO_4-silica. A small amount of water was incorporated into the reaction mixture after TLC results showed the creation of the di-O-isopropylidene derivative to hydrolyze the volatile 4,6-O-isopropylidene specifically. Pyridine was used to neutralize the mix, and the solvents were then eliminated. The residue was then again dissolved in pyridine before being benzoylated with BzCl. When the procedure was completed, the solvents were evaporated, and the precipitate dissolved in 80% AcOH. The residual 2,3-O-isopropylidene acetal had been hydrolyzed after stirring at 80°C in the resultant suspension for a few hours. After solvent evaporation and chromatographic purification, the required derivative (18) was obtained with an 81% yield, as shown in Figure 10 [40].

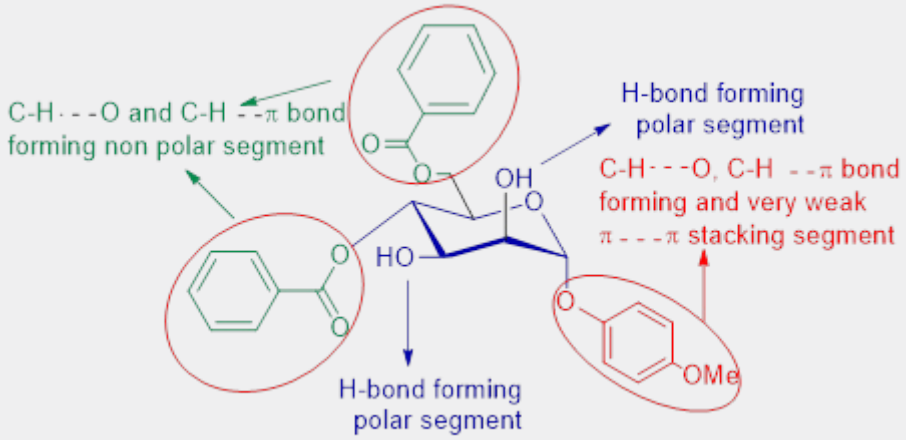

Figure 9: Mannose-derived carbohydrate-based gelator structure [40].

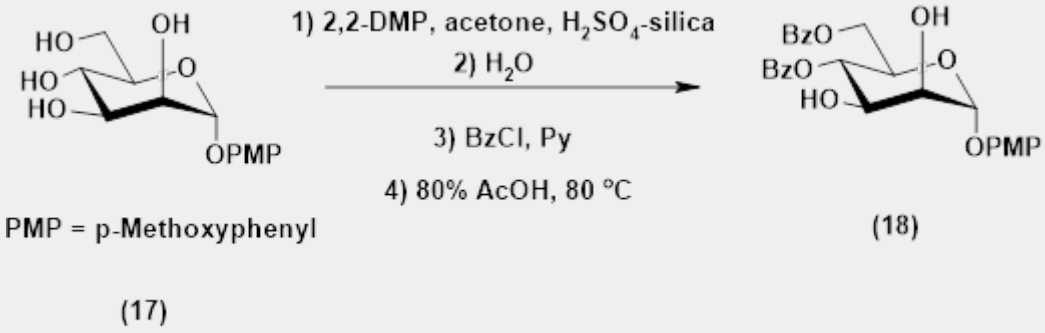

Figure 10: Synthesis of mannose derivative (18) [40].

Organogelators Based on D-Gluconic Acid

K. Fang et al. recently identified two effective and one-step-synthesized organogelators sustaining D–gluconic acetal derivatives. These developed from biphasic mixtures with water to exhibit phase-selective gelation behavior towards aromatic solvents [41]. The target gelators could be easily generated in a single step, as illustrated in Figure 11 It was worth noting that D-gluconic acid, a commercially accessible raw material, interacted with one equivalent of the corresponding aldehyde to produce the Gluconic Acid Acetals 19a and 19b. Organo-gelators based on D-gluconic acetal derivatives displayed aromatic solvent phase-selective gelation. 19b, in particular, can coagulate benzene, toluene, and o-xylene in powder form at room temperature with

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gentle stirring. Systematic investigations show that the gelation duration can be reduced to 10 minutes under certain conditions, and the aromatic solvent recovery rate can reach 82%. Additionally, the toluene and o-xylene critical gel concentration (CGC) values were less than 0.1 weight %, upgrading gelator 19b to the super gelator category. Additionally, even after several repetitions of the sol-gel cycles, the gelation ability remained constant, showing that these gels were completely thermally reversible [41].

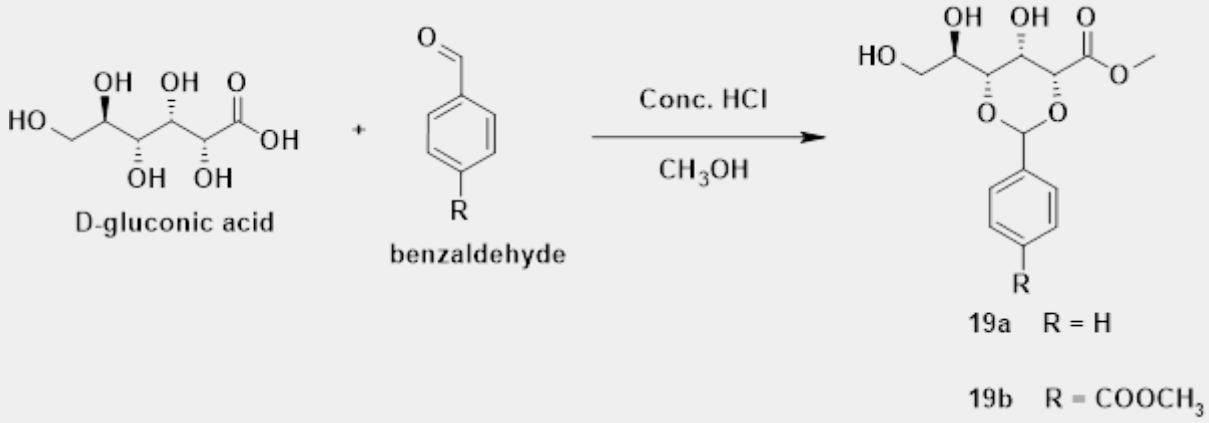

Figure 11: Synthesis of D-gluconic acid acetal derivatives 19a and 19b [41].

Arabinose Derivatives as Simple Organogelators

Two synthetically simple sugar-based super-organogelators (22c and 22d) with a partly ester-protected arabinose moiety were described. Because of the gelators' structural simplicity, very few interactions result in self-assembly during gelation. The gelators 22c and 22d were synthesized in three straightforward processes from commercially available D-arabinose (Figure 12). First, D-arabinose was transformed to mono-acetonide (20) by reacting with 2,2-dimethoxy propane using catalytic p-TSA. Diacetylation of compound 20 with acetic anhydride and pyridine in the presence of catalytic dimethyl aminopyridine (DMAP) yielded the 3,4-isopropylidine-1,2-diacetate derivative 21a. The chemical 21a was converted to compound 22c by treating it with acetic acid and water. Dibenzoylation of derivative 20 with benzoic anhydride and pyridine, followed by removal of the acetonide, resulted in the corresponding 1,2-dibenzoyl arabinopyranoside 22d as the β-anomer in the $^1C_4$ conformation [42]. Rheological characterizations of the gels show that the gels produced by the dibenzoylated arabinose gelator are mechanically more robust than those made by the diacetylated arabinose gelator [42].

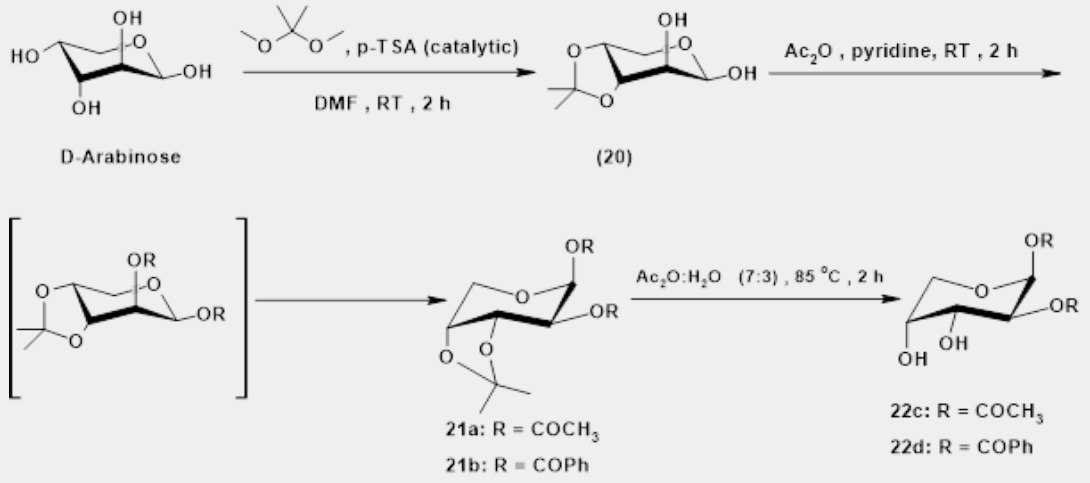

Figure 12: Synthetic route of D-arabinose derivatives 22c and 22d [42].

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Arabinose-Based Organogelators

Somnath Yadav and coworkers discovered two new triazolyl arabinoside gelators capable of gelling crude oil and other petroleum fractions and numerous hydrocarbon-based organic solvents and chlorobenzene. Commencing with commercially available D-arabinose, a series of triazolyarabinosides (23a-b) bearing different substituents on the triazole moiety adopted straightforward and high-yielding procedures [43].

The initial step was to convert D-arabinose to the per-O-acetylated derivative, then to the anomeric bromide using PBr_3, and finally to the anomeric azide via an easily performed nucleophilic substitution reaction by employing sodium azide. The above steps were reported using multi-gram scales before being followed by Cu(II) catalyzed "click reactions" with suitable acetylenes to generate the triazolylarabinosides 23a-b, as illustrated in Figure 13. Fundamental gelation analyses for the two compounds were reported in a wide range of solvents, and it was discovered that the two phenyl triazolyl derivative 23a and carbomethoxytriazolyl derivative 23b were effective organogelators for common hydrocarbon-based solvents that are organic [43]. The gelators described above are acidic, stable and water-insoluble, making them ideal for all types of crude oils. Using these gelators, crude oil's phase selective organogelator (PSOG) gives a practical and simple solution for resolving marine and terrestrial oil spills [43].

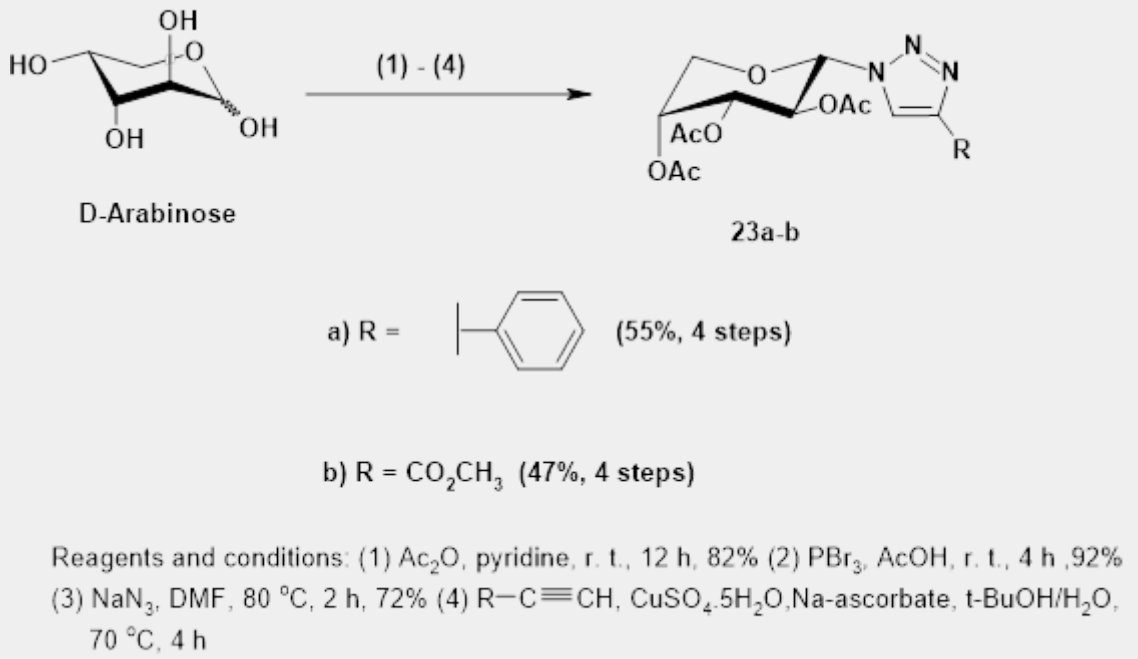

Figure 13: Synthesis of triazolyarabinosides (23a-b) [43].

Fabrication Of 1,5-Anhydrous-D-Glucitol, A Low-Molecular-Weight Organogelator Produced from Starch

Shiro Komba et al. produced low molecular weight organogelators, the derivatives of 1,5-anhydro-D-glucitol. This cyclic polyol results from starch's enzymatic and fermentation procedures, including different linear saturated fatty acid chains of various lengths using ester linkages. It was reported that these materials could turn distinctive organic gelatinous solvents. These gelators were created by warming powdery 1,5-anhydro-D-glucitol and powdery fatty acids to 230°C to melt the molecules without employing organic solvents or activated fatty acids and then finalizing the esterification reaction without or with a base catalyst. In this way, the two organogelators, 1,5-anhydro-2,3,4,6-tetra-O-palmitoyl-D-glucitol (24) and (25) with palmitic acid and stearic acid without including organic solvents or activated fatty acids in the chemical system were reported, and these structures were shown in Figure 14. This provided the fundamental technology needed for industrial mass synthesis. The gelators must be crystallized after the reaction to improve purity, and acetone was only employed at this point. The gelators described here can gel diisostearyl malate, which was used as a reference oil. As a result, producing these gelling agents at a meager cost is now possible [44]. The gelators were also demonstrated to be possible to generate without a base, resulting in whiter crystals compared to when a base was used. They determined that potassium carbonate is the optimal base for synthesizing 24 by evaluating yield, whiteness, and gel hardness [44].

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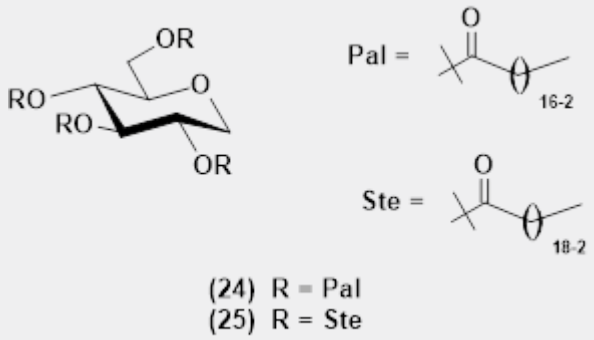

Figure 14: Organogelators, (24) with palmitic acid inserted into 1,5-anhydro-D-glucitol and (25) with stearic acid [44].

Polysaccharides -Based Organogelators

Polymers can be produced by living organisms throughout their whole life cycle. Plants, animals, bacteria, and fungi contain naturally occurring biopolymers. Proteins, nucleic acids, and polysaccharides are the most common biopolymers. The vast majority of naturally occurring polymers on earth are polysaccharides, which are macromolecules made of sugar units connected by glycosidic linkages. Natural polysaccharides are synthesized for a variety of purposes, including energy storage in plants, mainly starch, structural support of vegetal cells, namely cellulose, gelling agents forming the intercellular matrix and containing ions such as sodium, calcium, and magnesium that is alginate in brown algae. Some of them, such as starch, cellulose, polysaccharides from seaweeds and chitin, are commercially vital in various markets ranging from the manufacture of paper to food industry products [45].

Polysaccharides are biopolymers that belong to the family of carbohydrates. They are abundant in nature, low-cost to produce, biocompatible, non-toxic, bioactive, biodegradable, and water-soluble. Polysaccharides, for example, have recently gained popularity due to their valuable properties, particularly their gelling capability [46]. Polysaccharide gels are of tremendous interest for biomedical applications such as tissue engineering, wound and skin care, and controlled medication release. These materials can also be employed in agricultural and environmental technologies such as selective adsorption of hazardous chemicals, nutrient depots, and water retention. An effective technique for forming polysaccharide gels by covalent crosslinking employs "click-chemistry" reactions [47]. Xylan and xylan-based gels are not hazardous and compatible with life, and the polysaccharide that they contain is not digested in the stomach or intestine but can be eradicated in the human colon, which is an intriguing property for developing biomaterials for targeted drug release [48].

Biomedical Applications of Polysaccharides

Due to their biodegradability, non-toxicity, biocompatibility, and inexpensive processing costs, polysaccharides and their compounds are favored for medical purposes over synthetic polymers. Due to the advantages mentioned, polysaccharides obtained from natural sources are beneficial in the pharmaceutical, nutraceutical, food, and cosmetic industries. Currently, polysaccharides are employed in health care and to prevent illnesses, and an array of unique applications have also been found, including cancer diagnostics, inhibition, therapy, drug administration, anti-bacterial and antiviral approaches, and tissue engineering. Some biomedical applications of polysaccharides are Anti-Microbial, Antiviral, Anti-Tumor/Cancer, Anti-Obesity, Hypocholesterolemia, Anti-Diabetic, Gastro-Protective, Immune Modulatory, Anti-Inflammatory, Neuro-Protective, Antioxidant, Tissue Engineering, Wound Healing, Wound Dressing, Drug Delivery and Controlled Release [49].

Cellulose-Based Organogel

Cellulose (Figure 15) is the most prevalent biopolymer on the planet. All plants have cellulose in their cell walls, but it is also present in fungi, algae, and a few aquatic organisms of the tunicate's species and invertebrates. Cellulose is a linear homopolysaccharide comprised of d-glucopyranosyl units connected by β-(1→4) glycosidic linkages. Wood pulp and cotton fibres are the two most economically utilized cellulose sources. Cellulose has excellent film-forming capabilities and great chemical stability, and it is simple to produce cellulose derivatives. Besides cellulose derivatives, one of the most fascinating materials is cellophane, made from regenerated cellulose. Since cellophane is a thin, transparent material with high protective qualities against grass, oil, water, and bacteria, particularly since 1930, it has been used in commercial food packaging for fresh fruit, vegetables, sandwiches, cookies, and bakery goods [51]. And it has come into use in many practical areas, including its historical applications in furniture, clothing, and medical products [51].

By soaking five-weight percent cellulose/ionic liquid (1-butyl-3-methylimidazolium chloride, BMIMCl) solutions in liquids that are organic with the appropriate polarity, simple techniques for the production of cellulosic organic solutions and organogels were

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established. The right cellulosic solutions and gels were created when the 5-wt% cellulose/BMIMCl solutions were steeped in organic solutions with high and medium polarity (compared to permittivities). As illustrated in Figure 16, Cotton cellulose and BMIMCl, which had dried earlier under less pressure at 100 °C for three h, were combined. The resulting mixture was then allowed to remain at room temperature for 24 hours before being heated at 115°C in a vacuum oven for 3 hours to produce a 5-wt% cellulose solution. An organized was made by soaking the solution in benzyl alcohol for three days at a standard temperature. The produced gel was extracted from the medium and cleaned [52].

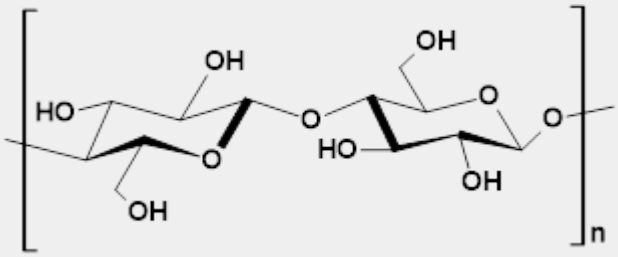

Figure 15: Cellulose's chemical structure [50].

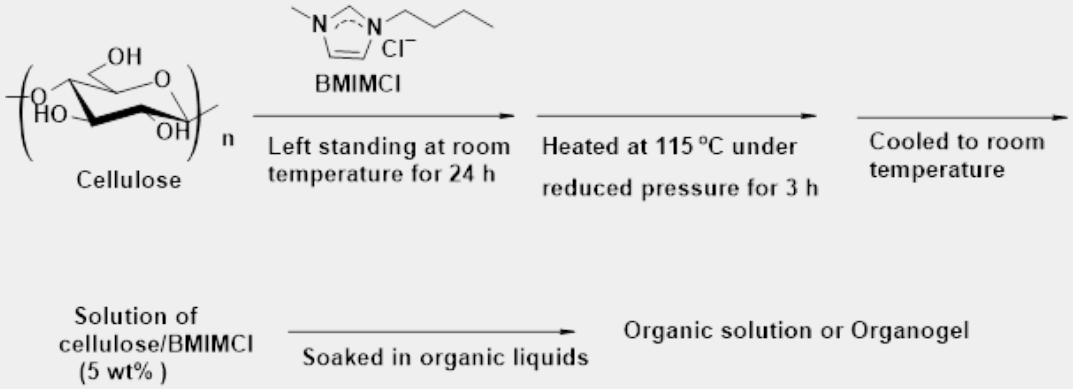

Figure 16: Synthesis of cellulosic organic solution and organogel by soaking cellulose/BMIMCl solution in organic solvents [52].

Chitin-Based Organogels

After cellulose, chitin (Figure 17) is the second most common polysaccharide in the world. It can be found in nature in many forms in crabs, insects, and mushrooms. It appears crystalline or semi-crystalline under a microscope, transforming this polysaccharide into a stiff and durable substance that acts as a barrier in the cell wall or cuticle and safeguards the organism [53]. Chitin is a historically structured amino polysaccharide that is the primary nanostructured component in a wide range of uni- and multicellular species' skeletal structures. Almost 200 years after its discovery, the chitin biosynthesis research in various phyla and its usage in healthcare, technology, and analogy is still essential. It is not easy to describe since it does not usually occur in its pure form, instead appearing as nano-organized chitin-proteins, chitin-pigments, or chitin-mineral composite biomaterials. Since the first chitin-producing prehistoric fungi appeared on Earth, chitin has been one of the primary structural biopolymers in the animal kingdom. Pure chitin has considerable resistance to alkaline treatment, one of its essential characteristics [54]. Units of D-glucosamine molecules and N-acetyl groups make up chitin, producing monomers joined by β (1→4) bonds. To be categorized as chitin, its degree of acetylation must be greater than 50 %. As a result, it is known as β (1→4) -N-acetyl-D-glucosamine [55].

It was demonstrated that chitin could be used as a filler ingredient to create biodegradable and biocompatible organogels, and triglycerides can act as a continuous hydrophobic phase. A substantial degree of aggregation was seen while using crude chitin, which hindered the creation of stable organogels. Two ways were adopted to reduce this aggregation and promote stability. Surfactants were employed to improve the dispersibility of the crude chitin, or the natural chitin was acid hydrolyzed into finer rod-like nanocrystals. For large chitin concentrations (20 wt%), both techniques developed constant organogels with preservation properties of up to 106 Pa [57]. Initially, precise amounts of freeze-dried chitin nanocrystals (ChN)/phosphatidylcholine or chitin-surfactants were mixed with water and triglyceride

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solvent (oil) using a high-speed blender for a few seconds. The dispersions were sterilized for 30 minutes on a stirring plate at 85°C. The organogels were then allowed to cool at ambient temperature before being kept in a refrigerator at 4°C. Regarding crude chitin or ChN, organogels were produced with chitin concentrations of 2,3,5, 8, or 20 wt%. Enzymatically altered phosphatidylcholine and ChN were consistently present in a 2:1 ratio with phosphatidylcholine in crude chitin samples containing 5, 10, or 15 wt% of phosphatidylcholines. Finally, organogels comprising ten wt% ChN and five wt% phosphatidylcholine or ten wt% crude chitin and five wt% phosphatidylcholine were treated with 5 to 25 wt% of waters [57].

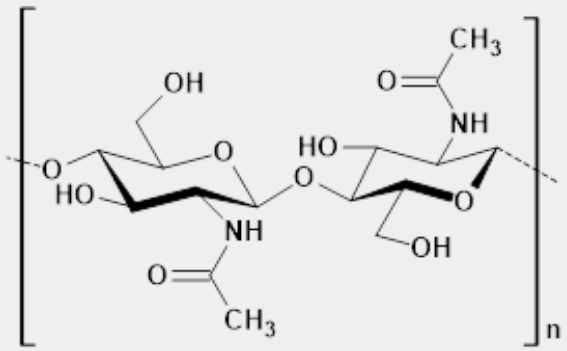

Figure 17: Molecular structure of chitin [56].

Curdlan-Based Organogels

Curdlan (CD) is a polysaccharide generated by Alcaligenes faecalis made of (1→3)-β-D-glucose units. When heated, CD produces a gel and is employed in cosmetics and the food sector. Moreover, CD hydrogels are acetylated to produce CD organogels, which have a peculiar swelling behaviour for continuous drying and swelling treatments. As demonstrated in Figure 18, the media was altered from water to acetic anhydride/pyridine after placing the hydrogels in ethanol. After that, the CD gels were treated with acetic anhydride and pyridine for three days at 70°C. The CD gels remained swelled in the medium used throughout acetylation and continued to pour in pyridine under dry conditions at room temperature. The resulting organogels were rinsed with chloroform to remove acetic anhydride and pyridine [58].

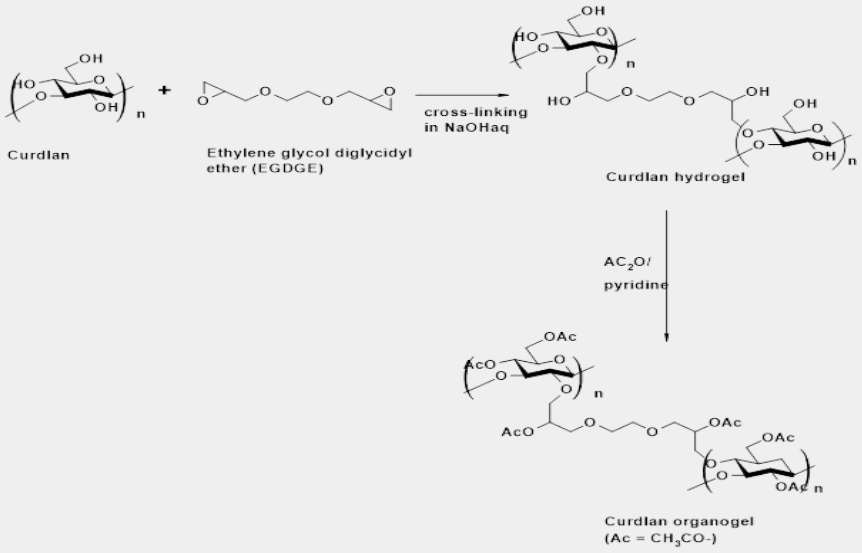

Figure 18: Covalently cross-linked curdlan hydrogel and organogel synthesis and representative molecule structure [58].

Organogels Synthesis Using a Diol-Containing Polyether Produced from The Sugar D-Xylose

Hannah S. Leese and coworkers recently described a unique organized system created by cross-linking a polyether derived from sugar with 1,4-Phenylenediboronic acid (PDBA). Cis-1,2-diols can be revealed along the polymer backbone by post-polymerizing the polyether generated from D-xylose. These hydroxy groups were incorporated to create dimethyl sulfoxide

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(DMSO) organogels with changeable rheological features and self-healing properties. Through post polymerization customization of a polyether made from D-xylose, a sugar-derived, diol-containing polymer was created, as shown in Figure 19. An oxetane-type monomer (26) was produced from D-xylose in three stages. A regioregular polyether (27) was produced by anionic ring-opening polymerization of compound 26 with potassium tert-butoxide and 18-crown-6 at a ratio of 100:1:1. Almost substantial deprotection of acetal groups was accomplished by acid hydrolysis, resulting in DMSO and water-soluble polyether's, 28 [59]. To create cross-linked organogels, the newly discovered hydroxy groups in polymer 28 could interact with difunctional boronic acids. A small amount of solution of 28 in DMSO was combined with a few equivalents of PDBA (about the deprotected monomer unit, considering quantitative acetal deprotection), as shown in Figure 20. A vial inversion test qualitatively showed that when stirred at room temperature, gel production took place in less than 5 minutes [59].

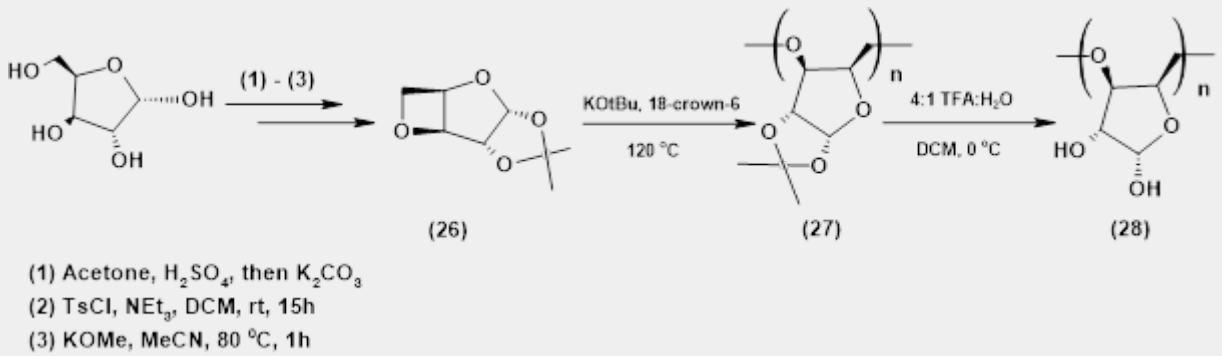

Figure 19: Synthesis of monomer 26, follow-up anionic ring-opening Polymerization, and post-polymerization acetal deprotection to produce the hydroxy-polymer 28 [59].

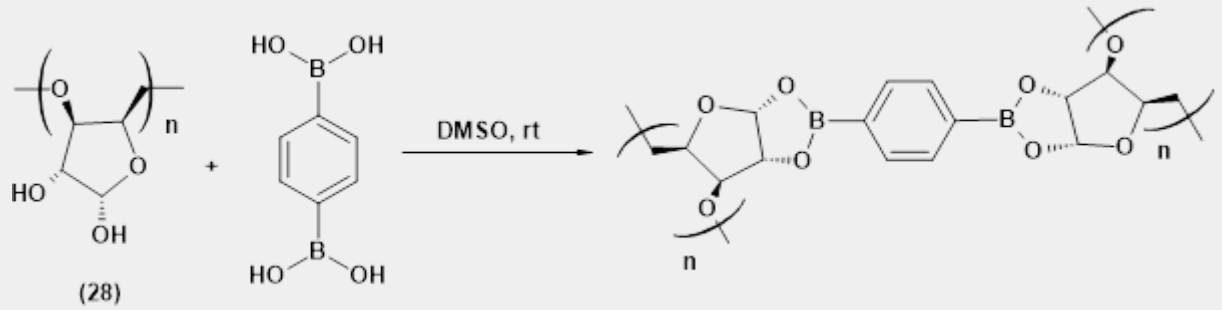

Figure 20: Cross-linking of 1,4- phenylenediboronic acid and polymer 28 [59].

Applications

Carbohydrates are essential in gelators not just as a hydrogen-bonding segment, but they may also be used to readily incorporate a variety of stereogenic centres into gelators by selecting from a saccharide library [52].

D-Sorbitol: A Low Molecular-Mass Gelator

Plants can produce D-sorbitol (Figure 21) from D-glucose and D-fructose chemically or enzymatically. It is mainly utilized in the food sector as a non-caloric sweetener and a critical intermediary in the synthesis of ascorbic acid. The essential attractive force responsible for the aggregation of LMOGs based on carbohydrates is thought to be H-bonding [60]. D-sorbitol in ethanol solution can be chilled and sonicated to generate a supramolecular gel. Many researchers altered D-sorbitol and reported the gelation properties of D-sorbitol derivatives in a wide range of solvents. Because it includes both hydrophobic and hydrophilic groups, the amphiphilic D-sorbitol derivative 1,3:2,4-di-O-benzylidene sorbitol can dissolve in various solvents. As a result, they can form strong gels in organic and polar solvents, such as polyethylene glycol and polypropylene glycol [61].

Meunier described the gelation phenomena of two benzaldehyde-D-sorbitols generated when a mixture of D-sorbitol and benzaldehyde was condensed with strong mineral acids in 1891, which was the first report of carbohydrate-based organogelators. Another group determined the chemical structure to be 1,3:2,4-dibenzylidene-D-sorbitol around fifty-three years later, a butterfly-shaped amphiphilic LMOG [33,62]. Figure 22

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shows the conversion of D-Sorbitol to 1,3:2,4-dibenzylidene-D-Sorbitol. Mannitol-derived gelator compounds have been demonstrated to have better gelating characteristics than other sugar alcohols like sorbitol and xylitol, whose structures are given in Figure 23 [61].

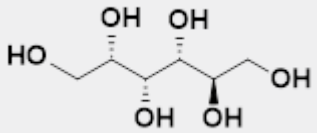

Figure 21: Molecular structure of D-Sorbitol.

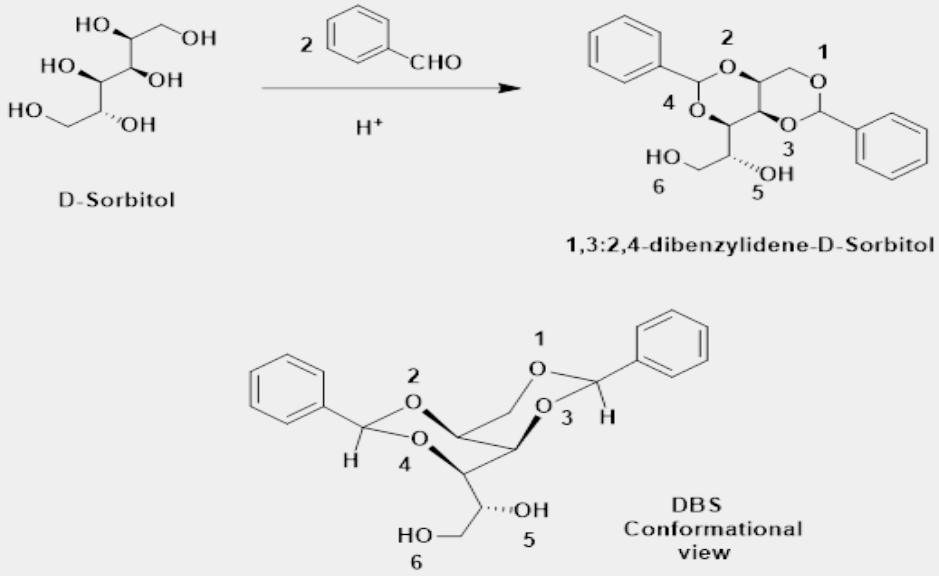

Figure 22: D-Sorbitol conversion to 1,3:2,4-di-O-benzylidene-D-Sorbitol and a conformational view of the structure of the molecule [63].

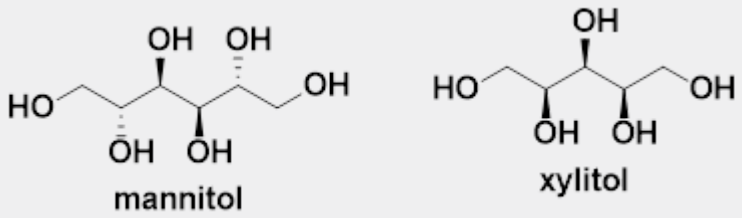

Figure 23: Structures of sugar alcohols, mannitol and xylitol.

Synthesis Of Polymeric Organogels Containing Beta-D-Glucose Pentaacetate by Reversible Addition-Fragmentation Chain Transfer (RAFT) Technique

In the presence of a di(ethylene glycol) dimethacrylate (DEGDMA) as a di-vinyl cross-linker, RAFT polymerization of mixtures of 2-hydroxyethyl methacrylate (HEMA) functionalized and acetyl (Ac) protected glucose derivative (Ac-glu-HEMA), which is a β-D-glucose pentaacetate containing methacrylate monomer produced carbohydrate-based organogels that could be converted to hydrogels by treatment with sodium methoxide. Solid-state ^{13}C cross-polarization/magic-angle spinning (CP/MAS) NMR, Fourier transform infrared (FTIR) spectroscopy, and field emission scanning electron microscopy (FE-SEM) was used to investigate these materials. The tendency of the organogels

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to swell was tested in various solvents with variable dielectric constants. DCM showed the most significant swelling ability, which also varied with the monomer/cross-linker ratio in the gel network. The adaptability and possibility of employing these gels for environmental and biological applications are emphasized [64].

Organogels From Benzohydrazide Derivatives

Different sugar benzo hydrazide derivatives that produce organogels with distinct morphologies in various organic solvents were created and analyzed. Twelve new categories of long alkyl chain-based hydrazide N-glycosylamine derivatives and their organogels have been identified [65]. Benzohydrazine and 4,6-O-ethylidene-D-glucopyranose, 4,6-O-butylidene-D-glucopyranose, and 4,6-O-benzylidene-D-glucopyranose were used to produce N-glycosylamine-based long alkyl chain substituted benzohydrazide derivatives (36-43) which are given in Figure 24. Through hydrogen bonding and van der Waals contact, it is discovered that the synthesized products can form stable gels in organic solvents. A known amount of glycosylamines was added to the solvent and boiled in a glass vial to obtain a homogenous solution. The vial was turned upside down after cooling to room temperature to confirm gel formation. Repeated heating and chilling proved the gelation's reversibility. The critical gelation concentration (CGC 1%) of benzohydrazide-N-glycosylamines was found by employing the smallest quantity of gelator required for gel formation at room temperature, and the gel research was performed using different solvents [65].

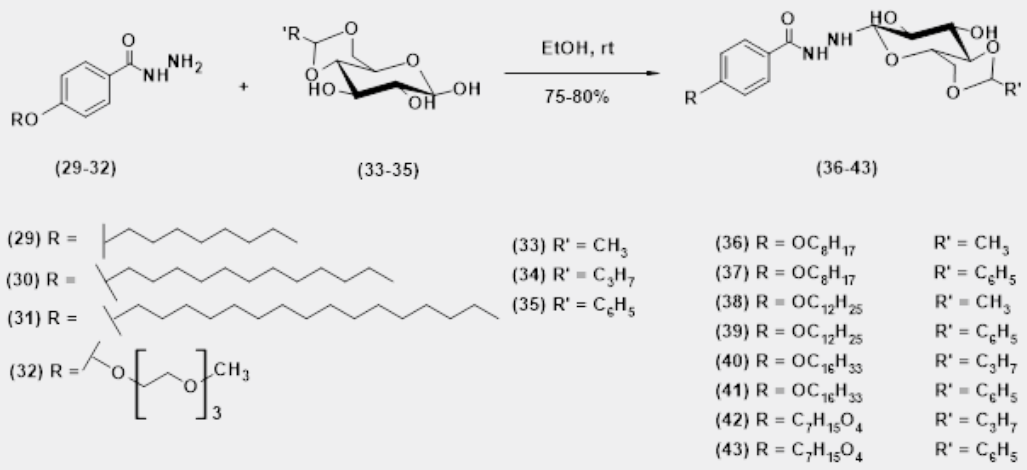

Figure 24: Production of long alkyl chain substituted benzohydrazide-N-glycosylamines (36-43) [65].

Carbohydrate Based Phase Selective Organogelators (PSOG)

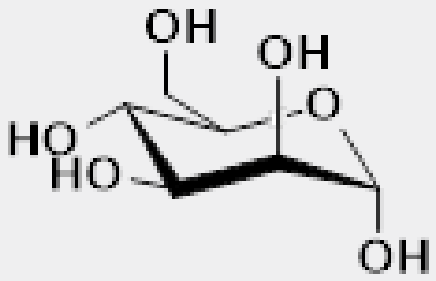

Figure 25: Structure of D-mannose.

Phase-selective gelators are organic compounds that may preferentially gelate the organic phase of a binary solvent mixture. Several phase-selective carbohydrates-derived gelators have been identified in the LMOG field. Potential low molecular weight organogelators from alkylated D-mannose (Figure 25) were produced and examined [66]. PSOGs have recently gained much attention due to their various roles in oil scavenging, dye removal, nanoparticle creation, etc. Also, they have been identified

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to eliminate watery pollutants, such as dye, effectively [67]. A glucose-derived Phase Selective Organo Gelators that can be applied directly as a solid without the use of a carrier solvent, as well as a feasible approach for oil-spill recovery that employs a low-molecular-mass organogelator, were developed by Amol M. Vibhute et al. These gelators were demonstrated to preferentially coagulate numerous oils, including crude oil, from oil-water mixes to create stable gels, which is a necessary attribute for effective oil spill recovery. Moreover, how these PSOGs can be included as a solid powder to an amalgam of crude oil and seawater, and then congealed oil can be extracted, as displayed in Figure 26 was shown by them [68].

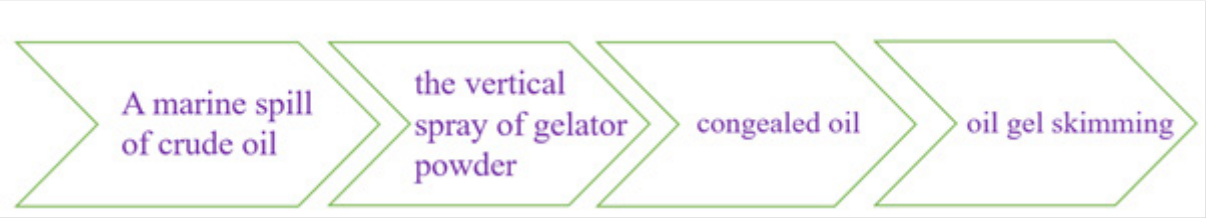

Figure 26: The process of extracting congealed oil by using PSOGs.

Organogelators From Carbohydrate-Derived Esters

The synthesis and characterization of fourteen 3-O-ester derivatives of 2-acetamido-2-deoxy-4,6-O-benzylidene methyl-α-D-glucopyranoside were carried out. Because this newly developed family of carbohydrate-derived esters can be synthesized using simple procedures using commonly available starting materials, they could have a wide range of practical applications, including oil spill cleaning. The systematic derivatization of N-acetyl glucosamine derivatives at the C-3 position resulted in an exciting class of low molecular weight organogelators. The overall structural characteristics derived from this series of compounds can be applied to different sugar derivatives, resulting in functional and diverse organogelators [69].

Diacetylene-Containing Galactose-Based Organogelator

A diacetylene-based galactose-based organogelator that was capable of congealing various organic liquids was developed. The gelator molecules are self-assembled so that the diyne motifs pile in a column, resulting in the topochemical polymerization of polydiacetylenes following photoirradiation. Because polydiacetylenes are semiconducting polymers, they have used this gelator to create polydiacetylenes-modified semiconducting fabrics, which are essential as antistatic fabrics and EMI shielding materials, by self-assembling the gelator on the fabric surface and then photo-induced polymerization to polydiacetylenes. The polydiacetylenes-cotton textiles demonstrated semiconducting behavior with a surface resistivity of 109 Ω/sq is a perfect resource for anti-static in nature or static dissipating fabrics [70].

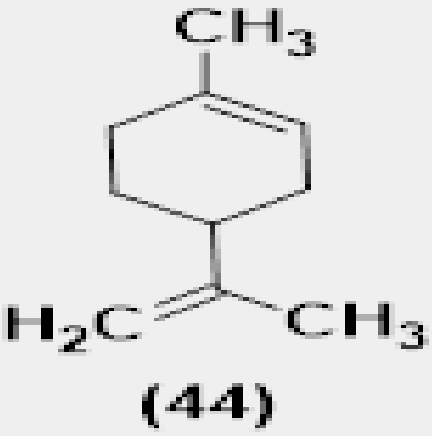

Figure 27: Structure of Limonene.

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Amino Acid-Based Organoglators

The basic biological building blocks, amino acids, may self-assemble into discrete nanostructures through supramolecular mechanisms [71]. The gel-to-sol transition temperatures (Tgs), which should be higher than the body temperature to maintain the gel state after administration, are crucial factors in determining the gel function. Limonene (44) may be categorized as an amino-acid-type gelator when it is included in dibutyllauroylglutamide in propylene glycol (PG) friendly organogels. In addition to the hydrophobic contacts between the lengthy alkyl chains, it has been suggested that these organogelators engage in substantial intramolecular hydrogen bonding among the amide groups in their structure. It creates a matrix made of solid fibres. It is made by combining dibutyllauroylglutamide, limonene, and PG, then incubating the mixture at 120°C. The combination cools down and turns into a white gel [17]. (Figure 27)

Organogels can load both hydrophilic and hydrophobic drugs. The physical characteristics of the organogels were enhanced by lengthening the gelators' alkyl chains [72]. There is much potential for safe in situ forming medication delivery since amino acids with various side chains have been widely employed to modify gelator structures to increase their biodegradability and biocompatibility [73,74]. The amino-based gelators always produce thermally reversible and long-lasting organogels. The injectable in situ-forming implant can increase patient compliance, increase medication retention duration, and lessen unwanted side effects. It is straightforward to administer. Drug delivery has extensively used organogels based on amino acid derivatives [75,76]. L-amino acid bioresources provide countless chances to create biodegradable and biocompatible polymeric materials due to their distinctive properties in terms of functional group variety, including carboxylic acid, amine, hydroxyl, aryl substitution, phenolic, and heterocyclic, among others [77]. The gel network for many organogelators comprises fibres, which then come together to form bigger aggregates. Usually, hydrophobic or van der Waals interactions cause cholesterol-based organogelators to cluster. Hendra M. Willemen and team created several new molecules (45) by coupling cholic acid to amino acid esters using diethylphosphoryl cyanide (DEPC) as a coupling reagent and amide bond formation. (Figure 28)

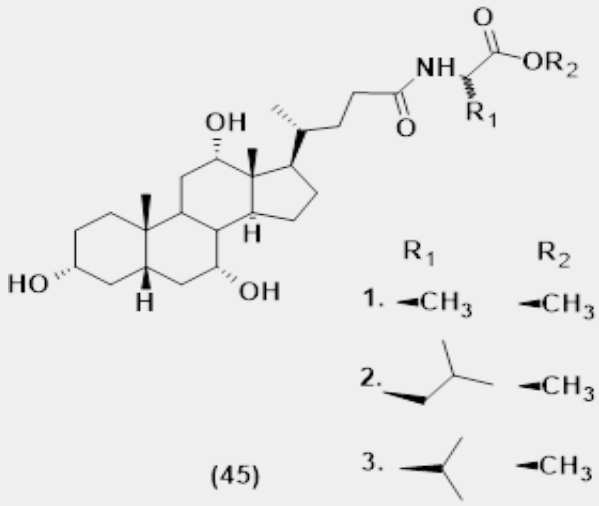

Figure 28: Coupling cholic acid with amino acid esters.

When cholic acid and an amino acid ester are combined, they serve as gelators for aromatic solvents, resulting in clear and stable gels [76]. Due to their innate biocompatibility, which increases their usefulness, amino acids and smaller peptide-based gelators are receiving increased attention among low molecular weight gelators [77]. Low-molecular-weight organic compounds that form gels in organic solvents are typically called physical or supramolecular gels. Organogels made by low molecular weight gelators contain a variety of nanoscaled superstructures, including nanoribbons, nanoparticles, nanofibers, bundles, nanosheets, and helical structures [78]. Mehmet Çolak et al. Synthesis N-alkanoyl-L-amino acid derivatives (46-49), Nε-alkanoyl-L-lysine derivatives (50-51) (Figure 29,30) To increase biocompatibility and gelation efficiency, lysine was combined with a variety of naturally occurring amino acids as well as with various alkanoyl groups. The fatty acids and amino acids used in gelators are chosen as biocompatible materials [78]. Suman Samai et al. have designed and synthesized the amide-baed organogelators (52-53) possessing six amide functional groups (Figure 31). The presence of these amide functionalities eventually resulted in the

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formation of amide-to-amide N-H····O hydrogen bonding between the molecules in suitable solvents and excellent aggregation. Dissolving in aromatic solvents produces stable gels with high mechanical strength. These materials are excellent for organo-gelating medicinal solutions [79].

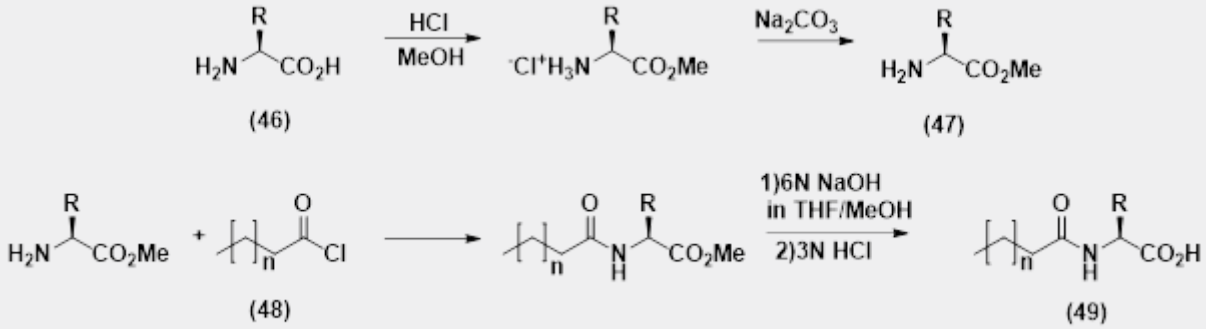

Figure 29: Synthesis of N-alkanoyl-L-amino acid.

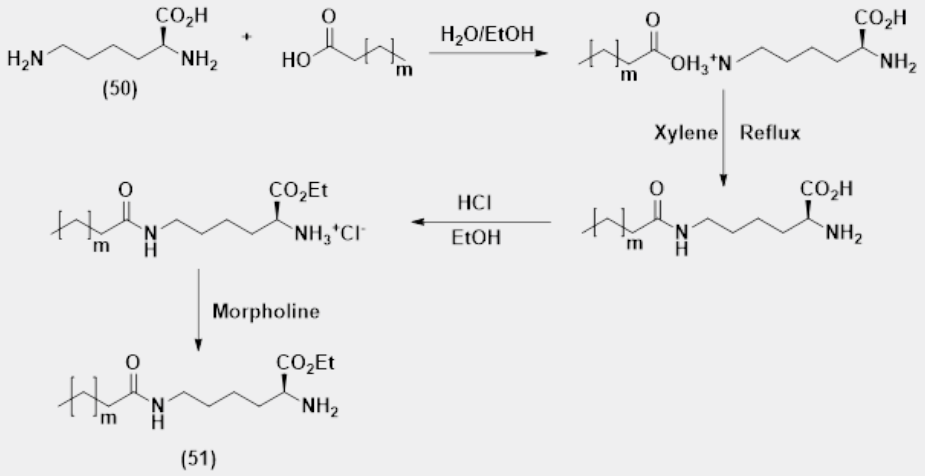

Figure 30: Synthesis of Nε-alkanoyl-L-lysine.

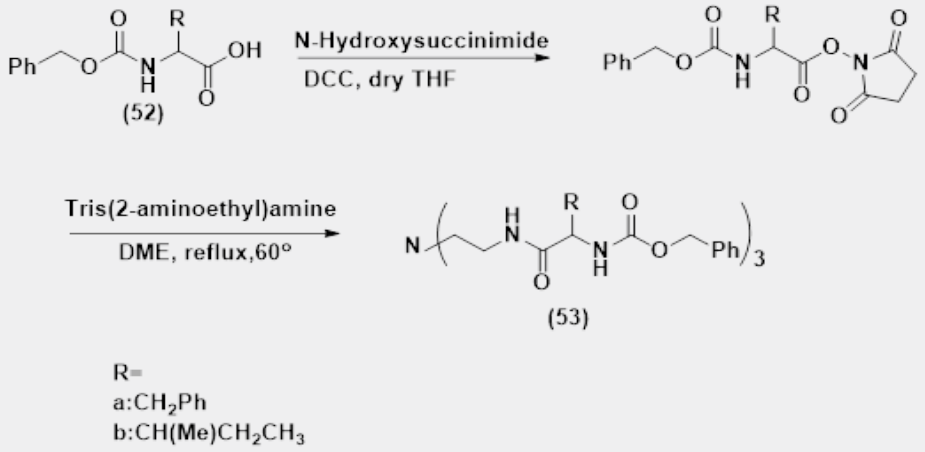

Figure 31: Synthesis of tripodal chiral ligand.

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Easy synthesis, customizable functionality, and low minimum gelation concentrations (MGCs) are appealing characteristics of LMOGs, making them desirable materials for applications including drug delivery, chemical sensing, and environmental cleanup. The MGC was found to be reduced by the straightforward structural alteration of adding a primary amide group in place of a terminal carboxylic acid [80]. Many LMOGs based on amino and fatty acids (FAs) have been developed to functionalize hydrogen bonding and van der Waals forces during gelation. Mridula Nandi et al. described an intriguing family of poly(methacrylate) homopolymers with phenylalanine (Phe) side chains attached that undergo self-assembly and gelation in certain organic hydrocarbons as a result of the connection between the side-chain functions. Because the phenyl group has a great propensity for -stacking interactions, it is well known in the literature that phenylalanine (Phe), a nonpolar hydrophobic amino acid, and its derivatives make remarkable low-molecular-weight gelators. (Figure 32)

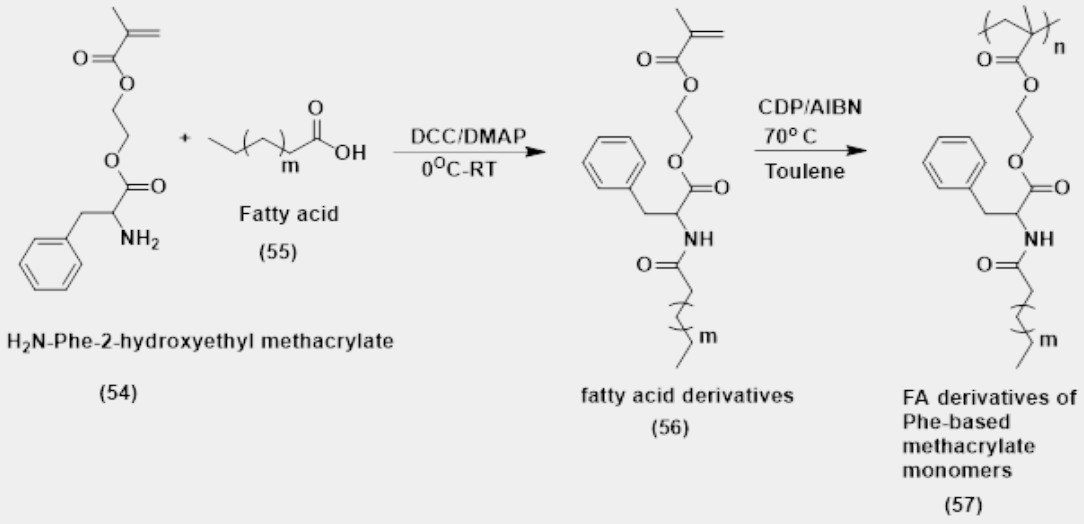

Figure 32: Synthesis of fatty acid-derived amino acid-based homopolymers (Phe-based methacrylate monomer) by reversible addition-fragmentation chain transfer polymerization (54-57) [81].

Selective aliphatic organic hydrocarbons, such as diesel, undergo macroscopic gelation due to a molecular self-assembly process driven by hydrogen bonding between amide groups in the side chains of the homopolymer. FAs and amino acids are commonly available, inexpensive, and biocompatible [81]. The ability of specific peptides and amino acids to self-assemble into fibrils has been used to create low molecular weight gelators. These hydrogels have potential uses in regenerative medicine, tissue engineering, and drug delivery [82]. Gelator molecules frequently form highly structured supramolecular groupings by molecular self-assembly, resulting in nanofibrils with micrometer lengths [83].

Long-chain hydrocarbons, amino acid derivatives, carbohydrate-derived systems, dendrimers, steroid derivatives, metal complexes, and even two-component systems are examples of the many molecular structures that make up these LMOGs [84]. Like in carbohydrate-base gelators, various interactions, including hydrophobic force, H-bonding interaction, conformational flexibility, and steric repulsion, worked together to create the three-dimensional fibrous network in the case of amino acid-based gelators [85]. Due to their abundance of hydrogen bonding groups and ease of modification with lengthy alkyl chains, amino acids are an ideal class of substances for creating such low molecular weight gelators. Furthermore, as α-amino acids are widely accessible, fatty acid amides of natural α-amino acids have undergone extensive research to form gel-type materials. It was shown that these α-amino compounds had good gelation properties for organic solvents like toluene and hexane. In addition to hydrogen bonding and the alkyl chain length, the gelation ability is influenced by other more subtle structural differences in the low molecular weight gelators, such as chirality. Branching due to the methyl group of β-amino acids prevented molecular packing, decreasing the intermolecular interactions [86].

PSOGs made of amino acids or peptides have received much research due to their nanofibrous structure that resembles collagen, high biocompatibility, robust molecular designability, and sensitive sensitivity to external field stimuli [87]. Marine oil spills pose a massive threat to the environment, causing ecosystem destruction, and fuel oil adulteration is also a significant issue. Phase-selective organogelators can create a gel in one solvent out of two immiscible solvents preferentially, making this gelator particularly beneficial for applications to separate a specific phase

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[88]. Biodegradable in situ gels include simple administration and affordable production. The in situ developing drug depot is injectable and has a low viscosity before injection. The system can harden to a semisolid after being injected. The molecular structure of the amino-based low molecular weight gelators has a notable propensity to establish H-bonding while keeping the property of minimal tissue toxicity, which has garnered particular interest in them [72].

Beibei Hua et al. synthesized a series of amino acid-based gelators (58) to explore the role of the gelator structure on functional properties, intended to establish a link between the molecular parameters with gel properties (Figure 33). By adjusting the gelator's three backbone substitutions, it was discovered that a complex interaction of hydrophobic forces, H-bonding interactions, conformational flexibility, and steric repulsion played a significant role in determining the gelation properties. This discovery proposed a theoretical framework for controllable drug delivery implants. Various interactions, including hydrophobic force, H-bonding contact, conformational flexibility, and steric repulsion, worked together to create the three-dimensional fibrous network. According to their structure, the three gelator replacements show equivalent molecular interactions. The n-alkyl chains of R_2 demonstrated strong van der Waals interactions that included lengthening the alkyl chain. Because of their architecture, the amino acid residues in R_1 were more complicated [89]. Two PSOGs based on L-phenylalanine derivatives (59-60) were prepared by Abdellatif et al. These are biodegradable, readily synthesized, and ecologically friendly. They could gel phase-selectively with water and several oils, including paraffin, sunflower, and olive. The results are highly promising and may address many concerns about oil leaks and spills in the Nile River and industrial oil waste [90]. (Figure 34)

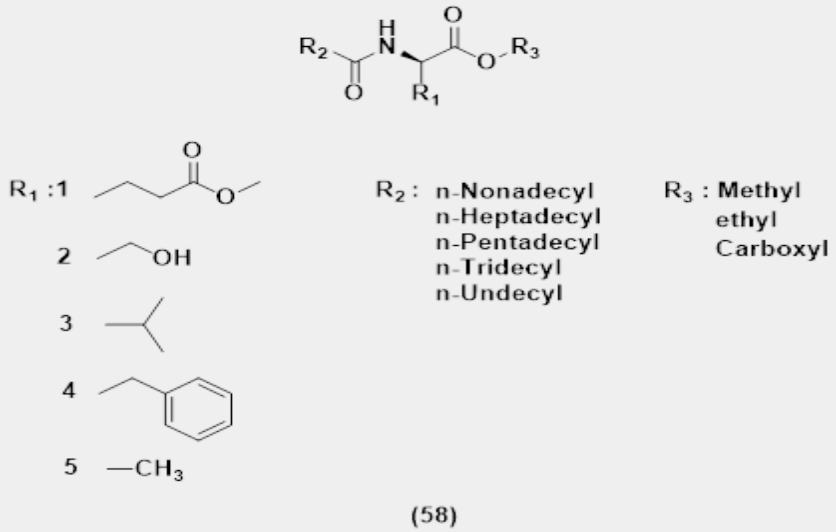

Figure 33: Structure of functionalized amino-based gelator.

A small amount of a biocompatible amphiphilic solvent, such as ethanol, N-methyl pyrrolidone (NMP), or dimethyl sulfoxide (DMSO), is added to the preparation before administration to make the organogel injectable at room temperature. Because this kind of solvent can also disrupt the interactions between gelator molecules, it is also known as an anti-gelation solvent. Beibei Hua, et al. designed and synthesized four new gelators based on L-alanine, L-glutamic acid, and L-serine (61). (Figure 35) The medications had a significant impact on the characteristics of organogels, such as a reduction in thermal stability with a lower transition temperature and a weakening of the organogel's mechanism [74]. Sonashree Saxena and Manickam Jayakannan combine the solvent-free melt polycondensation approach with the acetal-masking strategy in the L-aspartic acid system to design and manufacture new hydroxyl-functionalized polyesters (62) from L-amino acid resources. (Figure 36)

This polymer design may also be used in a wide range of other systems; it is not limited to specific cancer cell lines or medicinal molecules. Additionally, the hydroxyl functional groups might be investigated for attaching certain compounds or medications or used as the functional handle for cross-linking chemistry to make, among other things, hydrogels and organogels. Live-cell cellular absorption tests corroborated the potential of this novel hydroxyl polyester platform to display good cell killing in HeLa cancer cell lines. They confirmed the internalization of drug molecules at the intracellular level [75]. Sanjoy Mondal et al. synthesized several Schiff base-derived organogelators (63-66) (Figure 37) and have studied their gelation behaviour in various organic solvents, mixed organic solvents, fuel oils, and edible oils.

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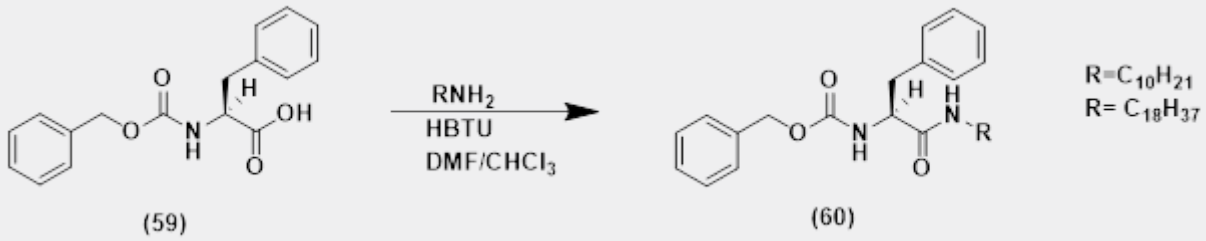
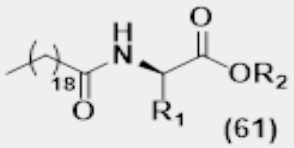


(59) (60)

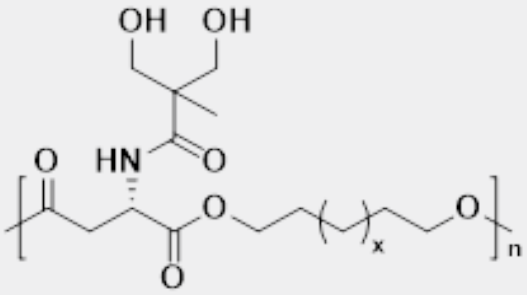
Figure 34: Synthesis of L-phenylalanine-based low molecular weight gelators [90].



(61)

	R_1	R_2
1.	Ehhoxyl	Methyl
2.	methyl	Methyl
3.	methyl	Ethyl
4.	2-dimethylglutarate	Methyl

Figure 35: Structure of amino-based gelator.



(62)

Figure 36: Structure of Acetal-functionalized L-Aspartic Polyester. x=4,6 and 8.

Among the gelators produced from Schiff bases, the one mentioned above (67) is appropriate for exhibiting quick, efficient, and selective gelation in the oil phase at room temperature, and gelator molecules may be recovered by straightforward distillation without causing the gelator to alter structurally. Within 60 seconds at room temperature, a solution of the above in THF applied to a water-oil combination using a gelator produces a gel promptly, effectively, and selectively in the oil portion of the mixture [88]. Suzuki, Hanabusa, and co-workers have used L-lysine as a critical building block to develop functional soft materials in organic solvents. Nε-Lauroyl-Nα-stearylaminocarbonyl-L-lysine ethyl ester (68) was first reported as an L-lysine-based organogelator, which can form long-term stable gels [91]. (Figure 38)

Four novel gelators based on β -alanine (69) that lacks chiral centres at the hydrophilic amino acid headgroup were created and characterized by Amrita Pal et al. However, the performance is less than gelators developed from L-alanine (70), which have a similar structural makeup. (Figure 39) Amrita Pal et al. concluded that depending on the molecular makeup of the gelator, chirality can either benefit or hinder gelation. The ability to gel is improved

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by the affinity between chiral head groups when the H-bonding interaction is weaker. If, however, the H-bonding contact is more significant, the latter effect amplifies it and causes crystallization [92]. Researchers have recently added a gelator as a structural agent to create a solid oil gel to prevent the trans-unsaturated fatty acids generated by inadequate edible oil hydrogenation. The range of structural agents in edible oils might be increased by using amino acid derivatives to solidify edible oils. Amino acids and their derivatives have been used in medicine because they tend to gel. N-lauroyl-L-alanine (71) and N-lauroyl-L-alanine dodecyl amide (72) amino acid gelators have been employed to create edible oil gels. Fan Zhang et al. explored the micro-rheological parameters and microstructure of these gels. (Figure 40) Fan Zhang et al. concluded that the rheological properties of gels prepared by N-lauroyl-L-alanine (71) and N-lauroyl-L-alanine dodecyl amide (72) considerably changed with the increase in gelator mass concentration at room temperature. The findings may help use vegetable oil gel and amino acid gel in food, medicine, and everyday chemicals [93].

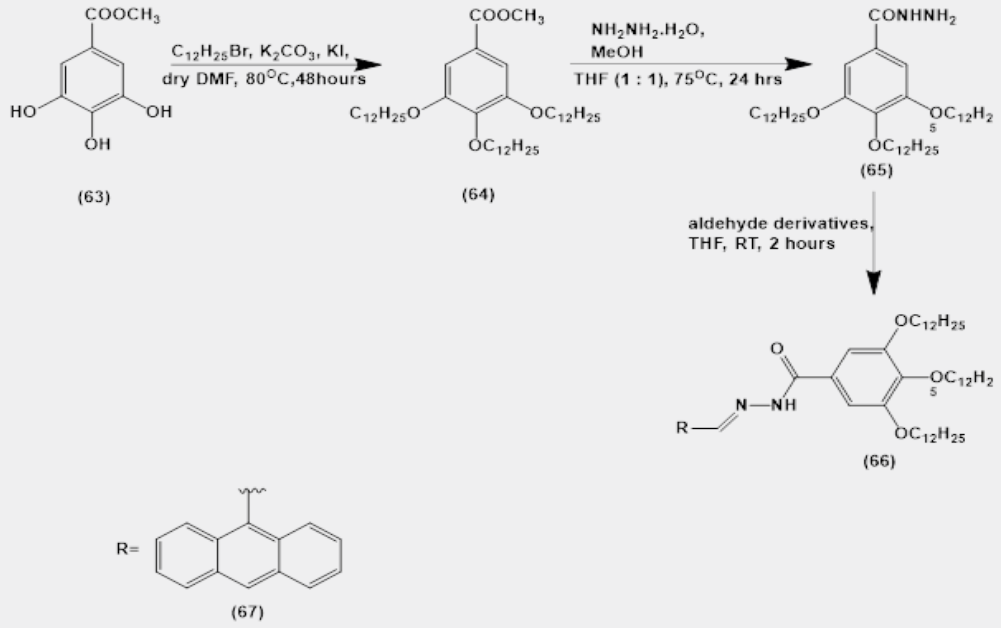

Figure 37: Synthetic scheme for producing Schiff base organogelators.

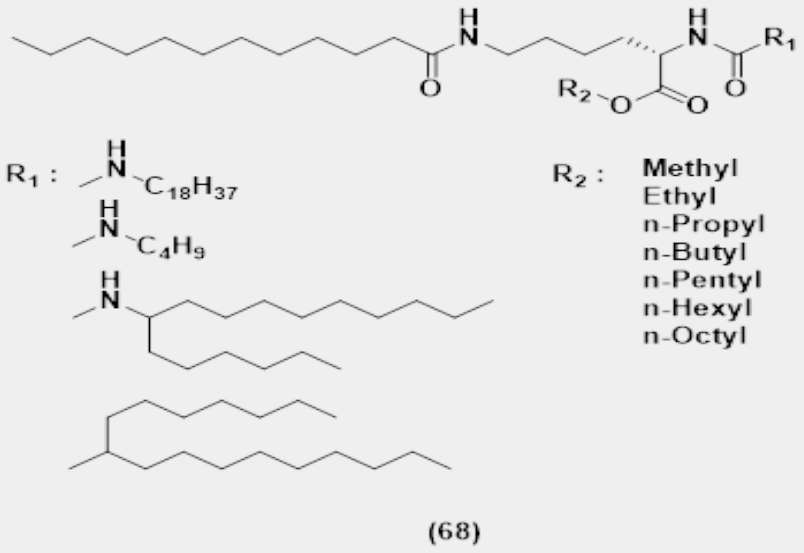

Figure 38: Structure of L-lysine-based organogelators.

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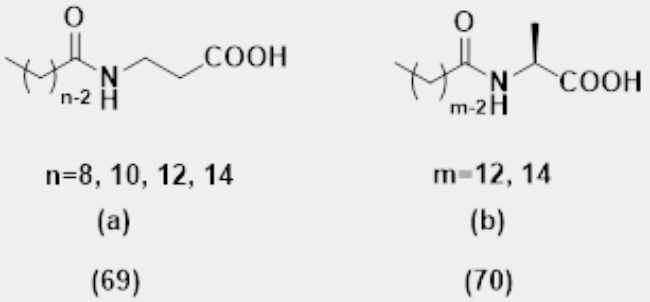

Figure 39: Structures of N-acyl- β-alanine and (69) N-acyl-L-alanine (70).

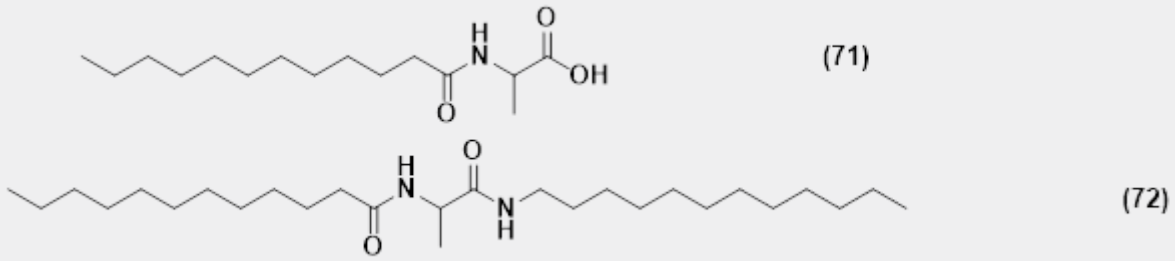

Figure 40: Structure of N-lauroyl-L-alanine (71) and (b) N-lauroyl-L-alanine dodecyl amide (72).

By derivatizing with aliphatic chains, Guillaume Bastiata and Jean-Christophe Leroux created biocompatible organogelator derivatives based on the aromatic amino acids like tryptophan, phenylalanine, and tyrosine (73). (Figure 41) Guillaume Bastiata et al. concluded that the derivative with the most extended chain length, i.e., $BTyrOCH_3$ (74), was discovered to have the most excellent gelation capabilities among the Tyr-based gelators. These Tyr-based gelators effectively deliver Alzheimer's medications via persistent parenteral infusion [94]. One of the most critical environmental issues now facing civilization is water contamination. Marine oil spills caused by the discharge of fuels and crude oil are a significant source of water pollution and a terrible environmental risk for marine ecosystems. The continuous use of organic dyes in several sectors is an essential cause of water contamination. Due to their high-water permeability, substantial adsorption surface area, ease of use, reusability, and superior biodegradability, hydro/organogel-based materials present an enticing alternative for removing colors from polluted water.

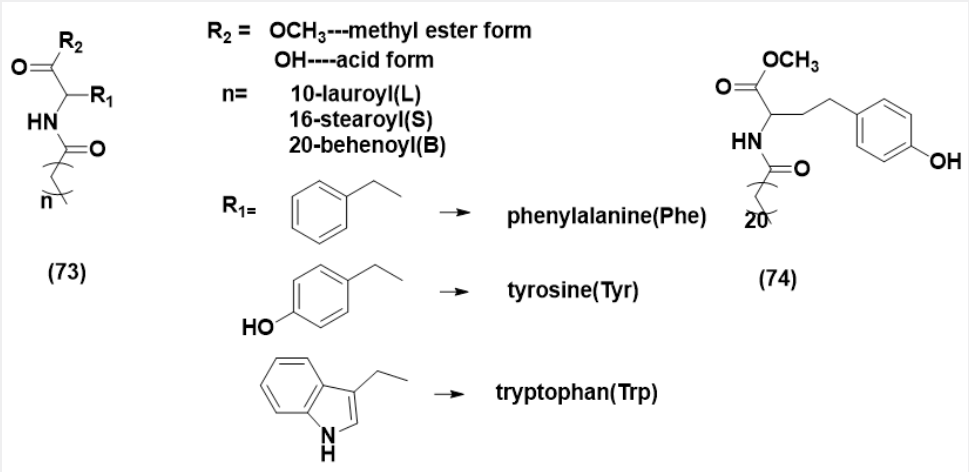

Figure 41: Structure of aromatic organogelators (73) and BTyrOCH3 Organogelator (74).

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Monikha Chetia et al. synthesized four dipeptides (75-78), ideal materials for oil spill recovery. They readily formed phase-selective, thermoreversible, mechanically robust organogels in many organic solvents and fuels such as kerosene, diesel, and petrol [95]. (Figure 42)

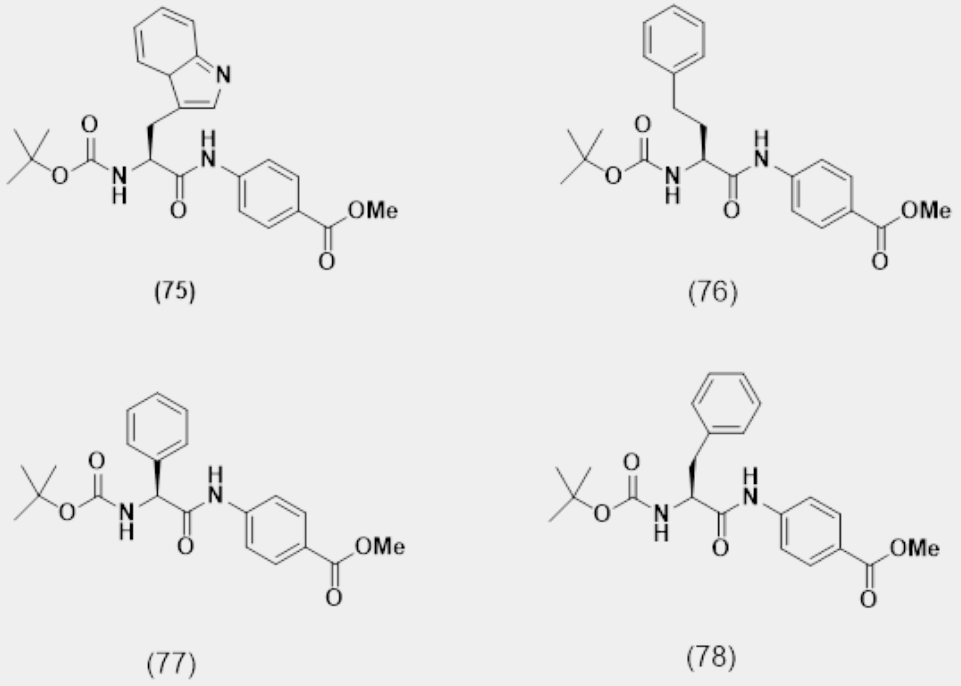

Figure 42: Structures of Dipeptide Low Molecular Weight Gelators.

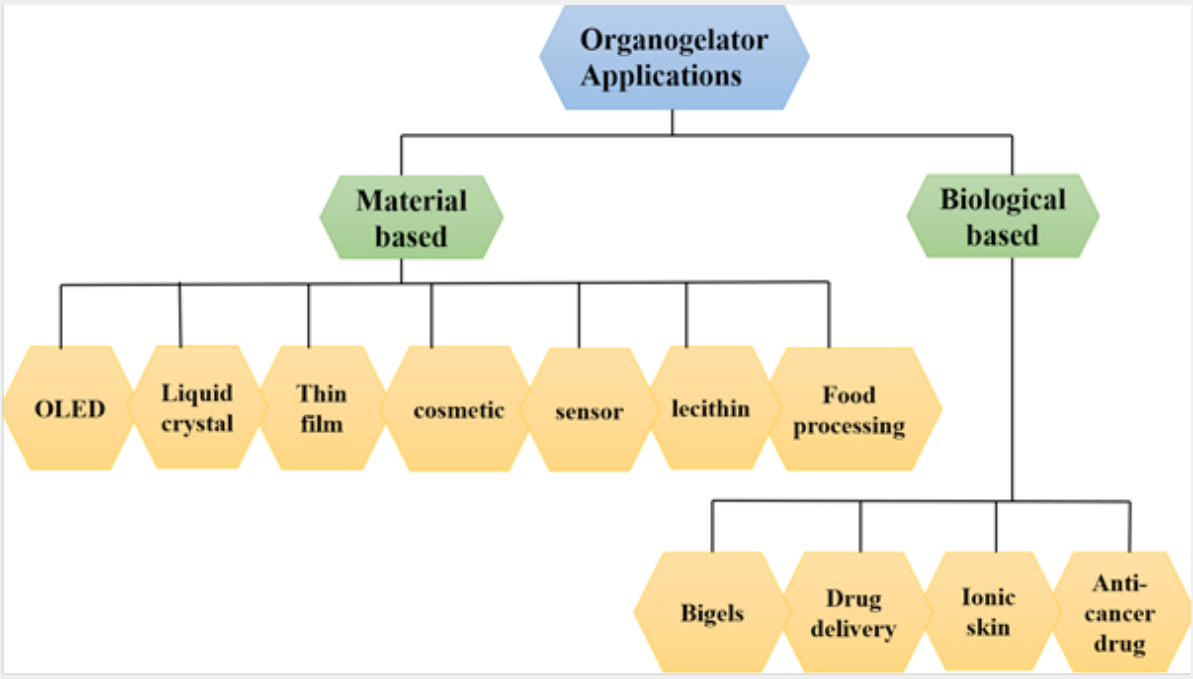

Figure 43: Flow chart on various applications of organogels.

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Material Applications of Organogels

Compared to hydrogelators, organogelators are reported to have broad applications due to some of the characteristic properties, such as weak molecular interactions and the responsive nature of the organic functional groups. Applications of these organogelators include the field of medicine, cosmetics, biotechnologies, and food science [96]. However, some organogelators are also used in biology, such as drug delivery, drug carriers, etc.; the following sections are limited to the applications of the organogelators in the field of material science and biology. Figure 43 depicts the various applications of organogels. The usefulness of organogels is in terms of component selection, unique features, and applications. Because of their unique properties, organogels have the most potential applications as complex multifunctional materials. The characteristics that distinguish this gel from others include multi-stimuli responses, affinity to a wide range of chemicals, thermal and environmental stability, electrical and ionic conductivity, and actuation [97].

Soft Material Organogrlators

LMOGs are a significant category of such materials being investigated extensively due to their distinctive physical properties and traditional structures. However, novel functional materials have been created due to recently expanded research in this area, providing new opportunities for organogels in OLEDs, sensors, cosmetics, and food industries, among other applications. Organogels are also among the most adaptable and promising types of multi-phase materials. According to recent articles, organogels fall into the following categories: i) "Apolar liquids, such as solvents made from organic matter or mineral or vegetable oils, are the continuous phase of organogels (or oleogels)" [18]; ii) "Organogels are soft materials made of polymeric systems that have swollen in organic solvents (hydrophobic polymer gels)" [99]; Furthermore, "Organogels are distinguished from hydrogels by adopting organic filling liquids" [98]. characteristics, aiming to highlight and promote organogels as a crucial class of functional soft materials. They also discussed the development of organogel applications. They also emphasized the significance of solvents in the functionality of these materials. Different polarities were essential for various applications to achieve optimal performance. Polar liquid media are necessary for biocompatibility, pH sensitivity, and ionic conductivity. Hydrogels address these applications but organogels with polar media can still be used (Figure 43) [97].

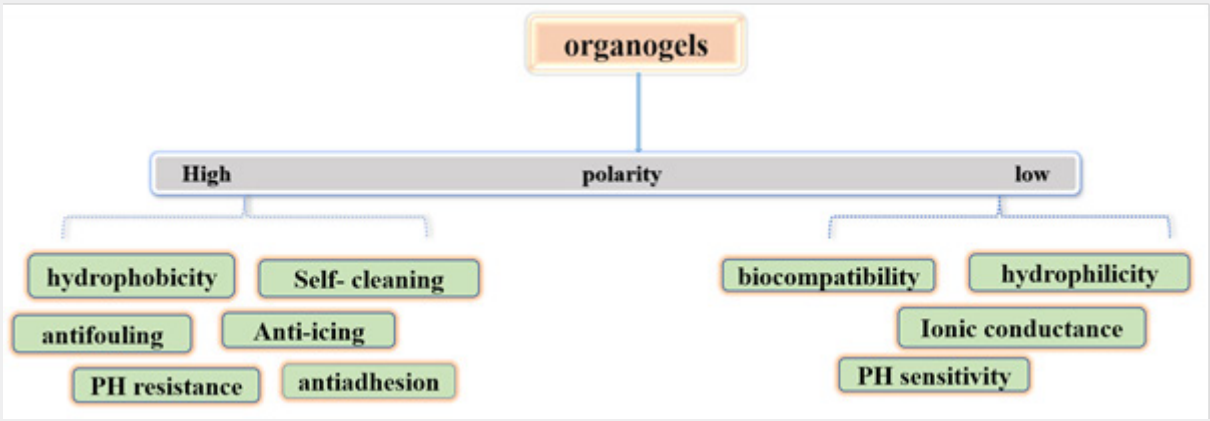

Figure 43: organogel applications based on polarity.

Zeng et al. study explains that organogel developments are focused on providing recent organogel progress worldwide. First, essential understandings of organogels are extensively summarized, including the elemental composition, gelation mechanism, production technique, and distinctive characteristics. A thorough investigation was done regarding various factors of the organogels, including the interaction between applied solvents and gelators, as well as gel behaviour and applications. They highlighted anti-icing, anti-fouling, droplet manipulation, drug delivery, food processing, and other applications [98].

OLED-Based Organogelators

OLEDs (organic light-emitting diodes) use organic materials to produce light. An OLED display's essential component is the emitter; When electricity is applied, it creates light as an organic (carbon-based) molecule. Due to their excellent qualities, such as colour display, high flexibility, low driving voltage, and quick response time, organic light-emitting diode materials have received much attention among material chemists. These characteristic properties enable their use in various applications, including next-generation flat-panel displays and solid-state

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lighting [99-102]. Small molecules and semiconducting polymers are now the two primary classes of organic chemicals used as light-emitting materials (EM) in OLED technology [102,103-105].

The research of Cristina Martin and coworkers describes the use of a self-assembling organogel, 5-(4-nonyl phenyl)-7-azaindole (79), as a novel emitter in small-molecule organic light emitting devices (OLEDs). Their studies suggest that monomer and dimer species coexist at high concentrations of compound (79). The presence of this type of dimer (produced via H-bonding) is responsible for the enhanced emission. The most striking characteristic, however, is the 3D network of enormously interwoven fibres generated in the organogel, which changes the photophysical properties. The electroluminescence bands deviate significantly from the photophysical properties, and a definite interrelationship between dimer formation and device performance was identified. Using a new self-assembly azaindole derivative as an emitter provides a new option for emitters in OLED applications (79), as shown in (Figure 44) [106].

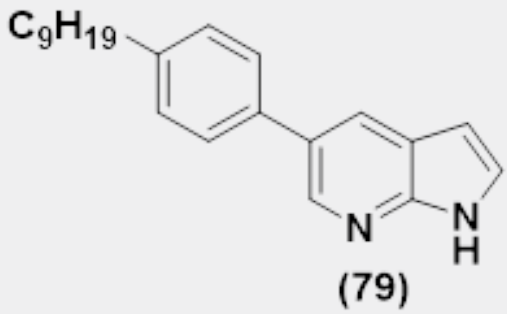

Figure 44: Scheme of compound (79).

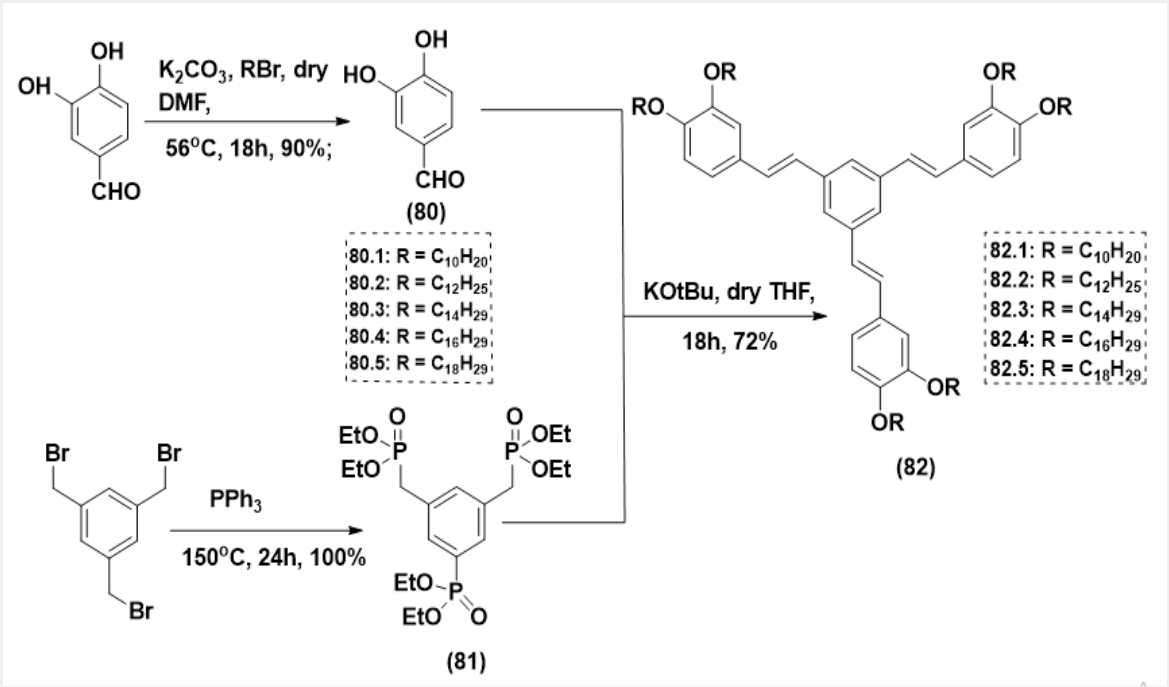

Figure 45: Synthesis of OPV derivatives (90). Reagents and Conditions: (i) K2CO3, RBr, dry DMF, 56°C, 18h, 90%; (ii) PPh3, 150°C, 24h, 100%; (iii) KOtBu, dry THF, 18h 72 %.

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De Joydip et al. has synthesized and characterized a new class of organic photovoltaic derivatives with thermo-reversible phase transitions from lam to Columnar rectangular when the peripheral chain length changes. All the compounds fluoresced blue in both the solution and solid states. OLED devices were made from compound 82.3, either as a host or as dopant-emitters in the N, N′-Dicarbazolyl-4,4′-biphenyl (CBP) host, at concentrations of 1, 3, 5, and 7 wt%. The device made with 1% yielded the best electroluminescent performance in terms of luminescence, current efficiency, power efficiency, and external quantum efficiency. To obtain the target molecules 82.3, the Horner-Wadsworth-Emmons reaction was carried out with 81 and 80.1-80.5 in the presence of potassium tert-butoxide in dry THF at room temperature under N2 atmosphere as illustrated in (Figure 45) [107].

Liquid Crystal-Based Organogelator

Liquid crystal is a state of matter with properties that fall between normal liquids and solid crystals. A family of gels known as liquid crystal organogels contains a liquid organic phase inside a three-dimensional, cross-linked network. Liquid crystalline polymers have received much attention because of their exceptional thermal and mechanical properties [108]. Yu Haifeng and the group worked, and a series of supramolecular liquid crystalline polymer organogels were effectively manufactured. The gelator was created using the supramolecular self-assembly of one azo-pyridine-containing polymer (83) and a carefully selected long-chain fatty acid (84), as shown in (figure 46) [109].

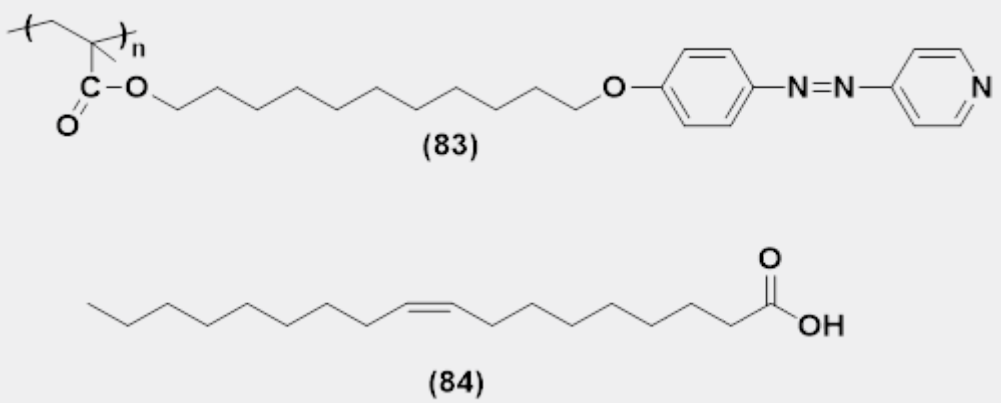

Figure 46: Chemical structures of the azo pyridine-containing polymer (83) and oleic acid (84).

The organogel's important phase transition in response to external triggers such as temperature, light, and Ag+ ion was thoroughly examined. Due to the inherent nature of the multi-responsive organogel, holographic gratings were recorded in the produced supramolecular liquid crystalline polymer organogel, displaying numerous switching behaviours to these stimuli [109]. According to G Baoxiang et al., Amphiphilic dendritic peptides G3 possess strong organogel properties with a minimal gelation value of one wt%. Over a wide temperature range, amphiphilic dendritic peptides G3 can also create a hexagonal columnar structure in a liquid crystal. Baoxiang Gao synthesized convergently using typical EDCI coupling of N-carbobenzyloxy-L-aspartic acid and L-aspartic acid dodecyl ester (previously made by esterifying aspartic acid with 1-dodecanol) and repeating a two-reaction cycle allowed for the progressive synthesis of the second (G2) and third (G3) generations in yields of 75 and 60%, respectively (Figure 47). The structure and purity of the amphiphilic dendritic peptides were confirmed using ^{1}H NMR, MALDI-TOF mass spectrometry, and elemental analysis [110].

B Subramanian and coworkers state that 1,2-bis[4-(4-(10-decyloxy) phenyl azo)]-benzoyl hydrazine (85), an azobenzene-based photo-responsive gelator with selective gelation in an alcoholic solvent, is presented. While polarised optical microscopy and differential scanning calorimetry are used to explore their thermodynamic behaviours, X-ray diffraction techniques assess the related layer arrangement, demonstrating a monotropic liquid crystalline phase in the organogel. The current work indicates that in the presence of an alcoholic solvent, the non-liquid crystalline gelator can be changed into a liquid crystalline organogel by disrupting the entanglement of gel fibres of self-assembly, resulting in a gel-sol transition, as shown in (figure 48) [111].

Thin Film-Based Organogelators

A thin film is a thin layer of material ranging in thickness from fractions of a nanometer to several micrometres. Many applications rely on the controlled production of materials such as thin films. Thin film organogelators comprise a liquid organic phase enclosed in a three-dimensional, cross-linked network. They are used to change surfaces by adding a thin layer of

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organogelator film that not only absorbs oil but also traps it in a crosslinked network, giving the material remarkable self-cleaning properties. This technique can be used with several popular engineering metals. Lei Jiang and coworkers developed self-cleaning organogel surfaces using free radical copolymerization of methacrylate monomers. Through an uncomplicated technique, they created organized-based easy-sliding covers with great self-cleaning. Because it does not use fluorinating reagents, this method is environmentally safe and may be used in popular industrial metals such as aluminium, copper, and iron. (Figure 49) shows the general design concept [112].

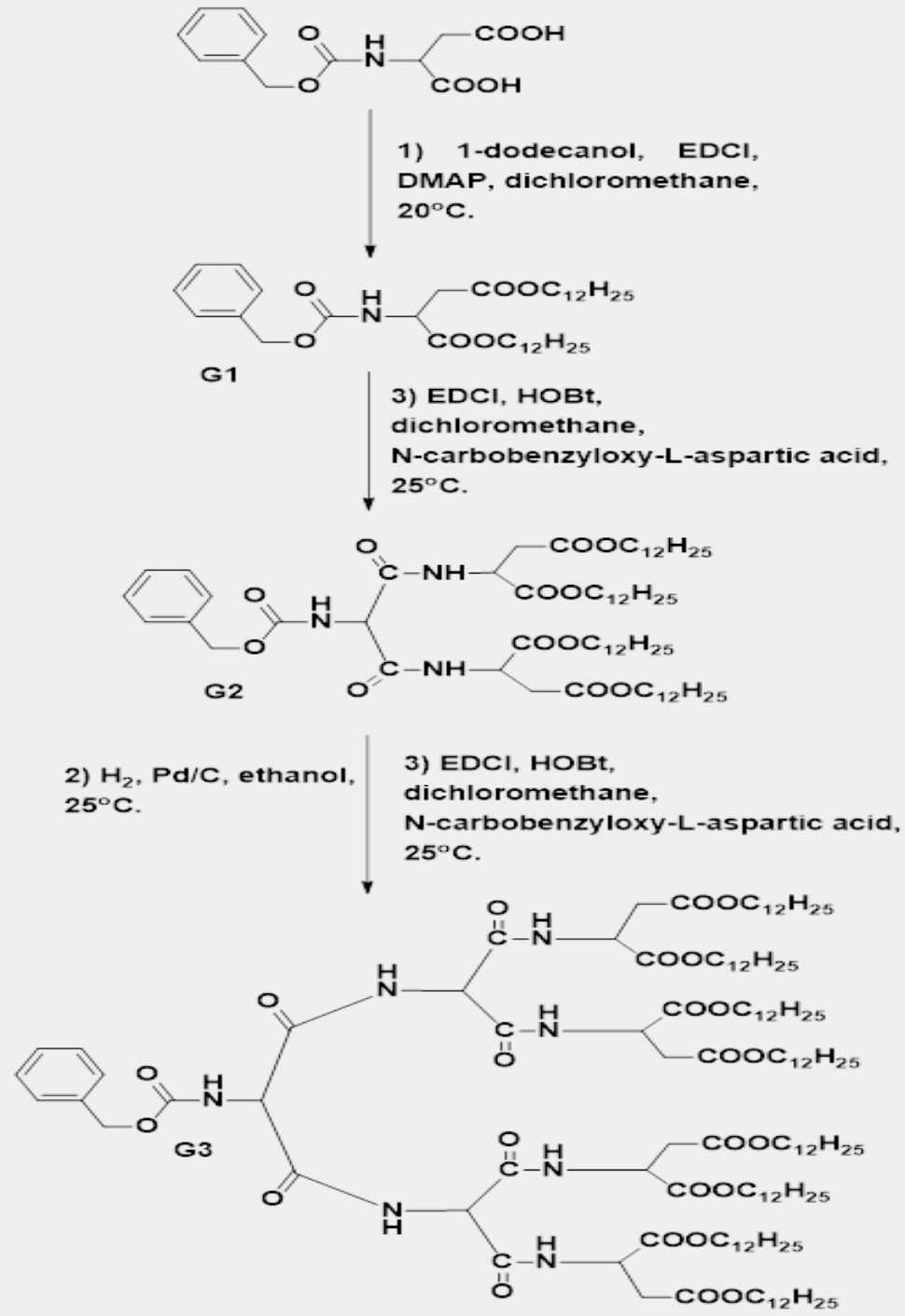

Figure 47: Amphiphilic dendritic peptides were synthesized in a specific order: (1) 1-dodecanol, DMAP, dichloromethane, EDCI, 25°C. (2) H_2, Pd/C, ethanol, 25°C. (3) EDCI, HOBt, dichloromethane, N-carbobenzyloxy-L-aspartic acid, 25°C. [110].

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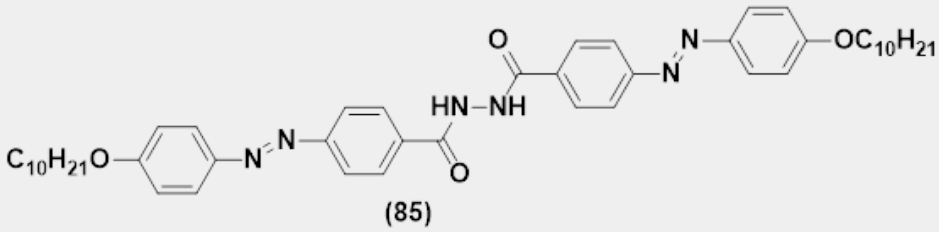

Figure 48: Molecular structure of 1,2-bis[4-(4-(10-decyloxy) phenyl azo)] benzoyl hydrazine (85) [111].

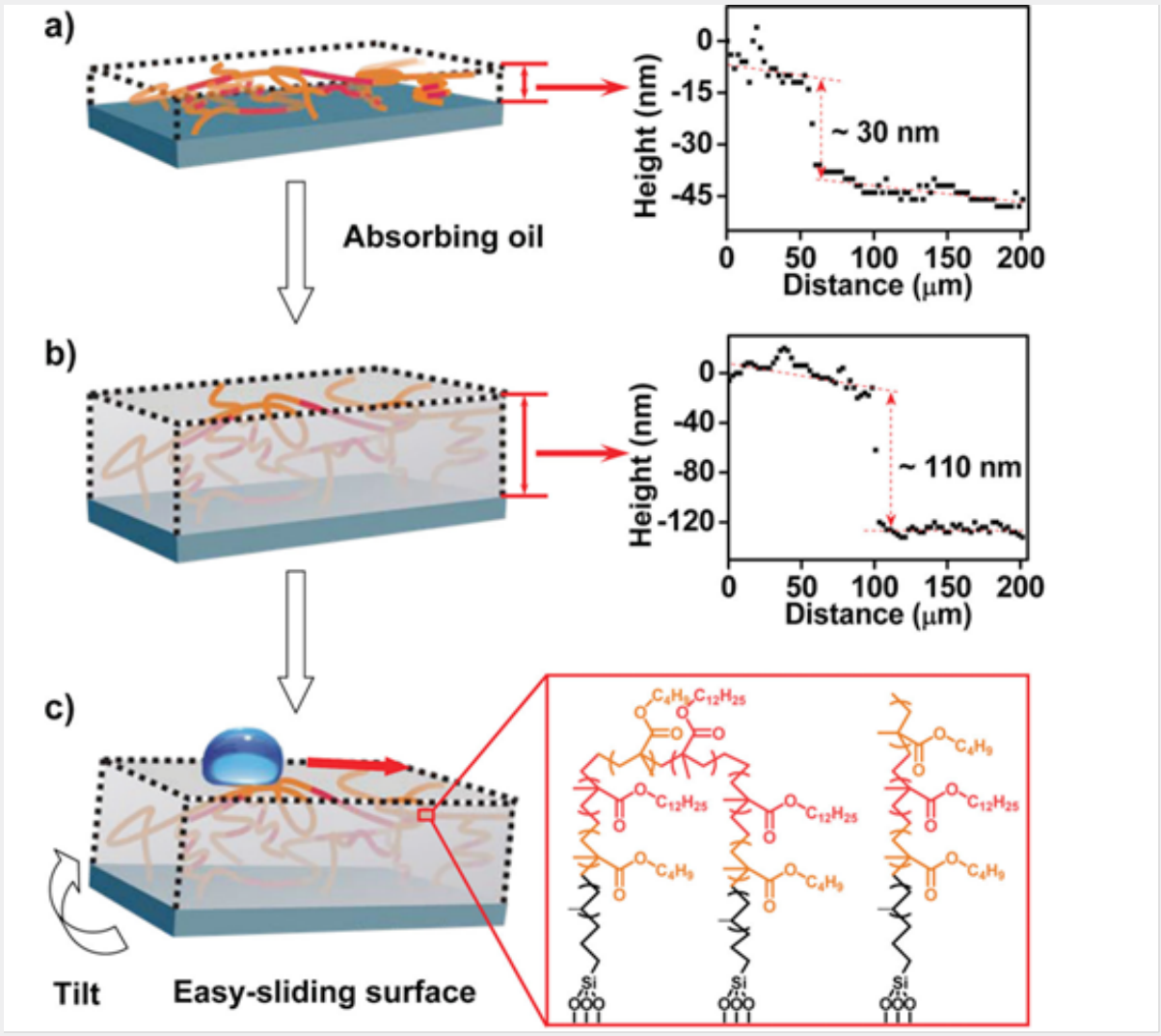

Figure 49: Surfaces made of organogel that are extremely self-cleaning. a) The organogel film on the surface has a thickness of around 30 nm. b) The thickness of the organogel film rose to about 110 nm after absorbing silicon oil (20 cSt). c) A water droplet can readily move across the surface by slightly tilting the organogel-modified surface [112].

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Cosmetic-Based Organogelator

Organogel cosmetic uses are still being studied. However, some benefits of employing organogels include low-cost composition, increased chemical stability of active compounds and physical strength of formulations, superior rheological qualities, and enhanced distribution profiles [113]. R. M. Martinez et al. investigated the characteristics and features of organogels and the benefits of using these systems in cosmetics. Cosmetic treatments improve skin appearance by utilizing vehicles with good sensory qualities. Several compounds exhibit organogelator activity. Lecithin, glyceryl fatty acid esters, sorbitan-derived esters, fatty acids, fatty alcohol, and vegetable waxes are some of the most common examples in cosmetics. (Table 1) discusses their applications [113]. C.L. Esposito, and Kirilov, P. report studies to assess the influence of low-molecular-weight organogelators (LMOGs), as a substitute alternative to waxes, on the thermal, rheological, sensory, and performance (textural and photoprotection efficiency) qualities of lipstick formulations. In vitro, sun protection factor investigations revealed that LMOGs, particularly the 12-hydroxystearic acid (12-HAS) lipstick formulation, significantly increase in vitro SPF and UVA-PF compared to wax-based lipsticks. LMOG-based formulations can replace some of the waxes in lipsticks due to their thermodynamic, mechanical, and photoprotective qualities [119].

Table 1: Organogelator used in cosmetics.

Organogelators	Uses	References
12-hydroxystearic acid	UVB blockers should be added to sunscreen to make them more stable.	[114]
Monoglycerides of fatty acids	The hydrophilic and the lipophilic templates.	[115]
Pluronic–lecithin	Deliver Centella asiatica to Cellulite treatment.	[116]
A mixture of glyceryl stearate and policosanol	cosmetic molecules delivery	[117]
Lecithin	Delivery of caffeine to cellulite treatment.	[118]

Sensors-Based Organogelator

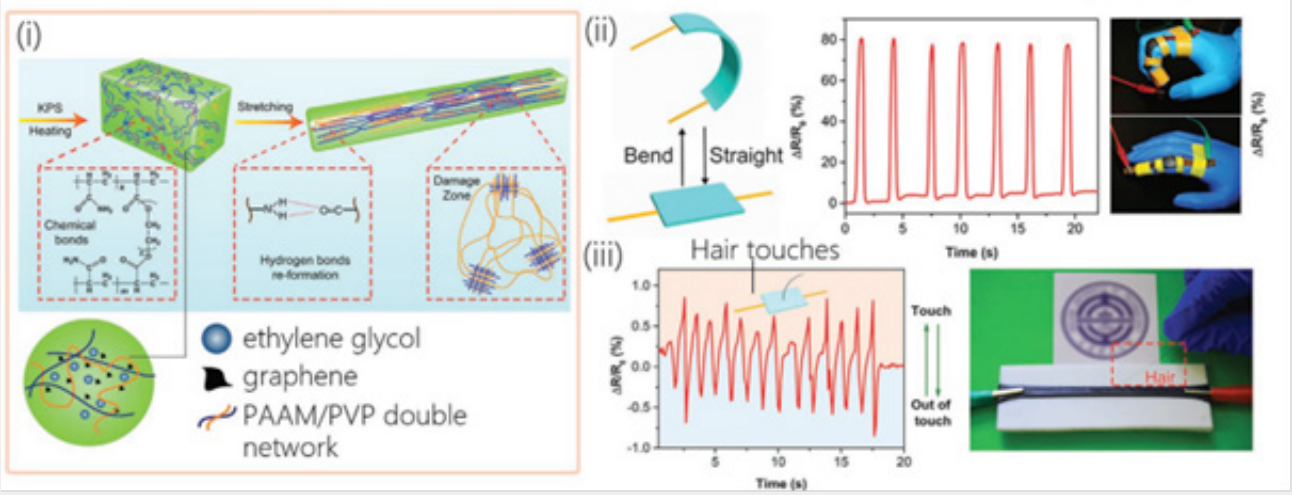

Figure 50: Highly stable double-network-based ethylene glycol organogel i) shows the reliable detection of joint movements, such as finger bending ii) and extreme sensitivity, as demonstrated by detecting hair touches after stretching 2.5 times iii). Reproduced with permission [122].

Organogels have been utilized to create sensors in which the properties of organic solvents play a role in sustaining, altering, and improving sensing performance. Thus, organogels can be loaded with tiny organic compounds with detection or sensing capabilities. Liu et al. demonstrated an organogel based on ionic polyacrylamide (PAAm) that is a sensitive electrochromic sensor [120]. Zhang et al. also demonstrated solvent-enhanced fluorescence behaviour on organogels based on triphenylamine-substituted acyl hydrazone derivatives [121]. H. Zhang and coworkers created an organogel with high stretchability (up to 10500%) based on a polyacrylamide-polyvinylpyrrolidone double-network in EG, including graphene. The generated

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organogel was utilized to detect motions ranging from significant e.g., i) thumb bending, ii) to gentle (e.g., inhaling) and tiny contacts of the stretched skin single hair, iii) ethyl glycol as a swelling agent enabled the uniform dispersion of graphene and thus stable conductivity throughout the gel; it also contributed to the sensor's stretchability as shown in (Figure 50) [122].

Lecithin Based Organogelator

Lecithin organogels are three-dimensional chains of self-assembled gelators, including an immobilized organic liquid phase. They are semi-solid structures. Because they have oil and aqueous formulation qualities, lecithin organogels are potential carriers for delivering a wide range of chemicals via the skin [123]. Lecithin organogels are gels composed of organic material in the liquid phase. Lecithin organogels are lecithin-based gels that are thermodynamically stable, transparent, viscoelastic, biocompatible, and isotropic (Figure 51), an organic solvent, and a polar liquid. Lecithin organogels have a jelly-like structure composed of three-dimensional networks of entangled reverse cylindrical micelles that immobilize the continuous phase, converting it from liquid to viscous gel [124]. Researchers have developed a wide variety of organogels and classified them according to the nature of the organogelators, such as Lecithin organogels, gelatin-stabilized organogels, limonene GP1/PG organogels, and non-ionic surfactant-based organogels, polyethene organogels [17].

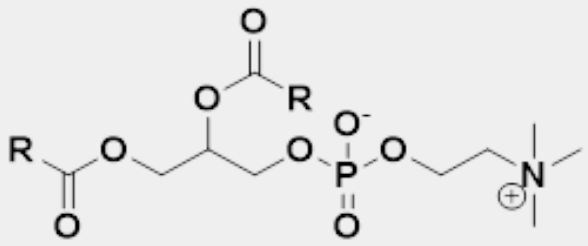

Figure 51: Structure of lecithin.

R. Sushil et al. investigated the ability of Lecithin organogels to deliver bioactive compounds for the treatment of skin ageing. Lecithin organogels offer a novel approach to the topical administration of antiaging drugs. Many good physicochemical features of Lecithin organogels are necessary for topical vehicles. It can dissolve hydrophilic and lipophilic medicines, making it a good vehicle for delivering a wide range of medications over the skin [118]. Bhatia and coworkers have conducted research on the many properties of Lecithin organogels, beginning with material selection and progressing to optimizing influential components and Lecithin organogels-specific characterization. The study's findings were intriguing, demonstrating a solid reliance on Lecithin organogels characteristics on the type and amount of phospholipid, Poloxamer™, auxiliary gelators, and organic solvent. The optimized Lecithin organogels were discovered to be relatively stable, easy to apply, and biocompatible [125].

Food Processing-Based Organogelator

Organogels have been explored for several years in food applications, including stabilizing water-in-oil emulsions and the controlled release of medicinal and nutraceutical compounds. In the food industry, applications include the potential use of structuring agents to reduce the migration of liquid oil into food, such as chocolate filling, margarine, baking products such as biscuits and cookies, puff pastry, and spreads, and to structure edible oils, reducing the use of Saturated fatty acids (SFA) and Trans fatty acids (TFA) [126, 127]. Even though organogels are semi-solid structures in which organic liquids (liquid oil) are restrained by self-assembled fibre networks, the entire system acts as solid fats. It possesses rheological and appearance characteristics that resemble fats. Siraj et al. performed a comprehensive investigation on organogel uses in processed foods highlighting the potential of these systems for the transportation of nutraceutical components, low-calorie emulsions, creams for toppings and fillings, spreads, lipid bases for baking products and comminuted meat products, among other things [128].

T Moschakis and group's research study Organogel emulsions of sunflower oil containing -oryzanol and phytosterols were created to manufacture frankfurter sausages with a partial bacon replacement. There were no alterations in pH, oxidation, or textural profile of links due to the addition of lipid gels. Organogels could partially replace bacon without appreciably changing the product's physical, chemical, or sensory qualities [129]. Organogels were created to replace the lipid phase of ice creams, lowering the SFA level. As an emulsifier, blends of 10% wax (candelilla, rice, or carnauba), 90% high oleic sunflower oil, and glycerol monooleate were tested. Compared to ice cream generated with high oleic sunflower oil, the quality of ice cream produced with rice bran wax improved; also, the organogel obtained with rice bran wax showed the capacity to substitute saturated fat in ice cream. However, when rice bran wax organogel is utilized as the source of fat, a high fat concentration (15%) and the glycerol monooleate emulsifier appear necessary to achieve

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a better ice cream structure [130-131]. Mert and Demirkesen's research studied the possible application of organogel from carnauba wax and candelilla wax to replace fats in cookies. Integrating 2.5 and 5% of the polishes in sunflower oil produced better-looking soft cookies with a lower consistency than regular fat. The lipid composition analysis revealed that organogels had higher quantities of unsaturated fatty acids than traditional fat, showing its potential as a healthier alternative for use in baked items [132].

Biological Applications of Organogels

Organogels have extremely adaptable qualities made possible by the inherent benefits of diversity in the gel components. The development of Organogel solvent components has been substantially aided by manipulating compound solvent systems, particularly nonpolar-polar organic liquid mixtures and polybasic organic liquid systems. Organogels retain the fundamental features of gels in a wide range of operating temperatures because of the organic liquid phase's tunable freezing point and boiling point [133].

Drug Administration

Drug administration is termed as distributing bioactive substances in dynamic human situations to achieve a desired treatment. Gels have an unmatched advantage in drug delivery due to their exceptional softness, biocompatibility, and bioactivity, notably, the entrenched networked nano/microstructures that serve as the foundation for pharmaceutical compounds. The main advantage of adopting this type of delivery route is avoiding the unpleasant gastric environment and first-pass metabolism. Organogels may be essential in the development of such delivery methods. In rat models, rivastigmine, an acetylcholinesterase inhibitor, has been successfully delivered subcutaneously using organogels made of tyrosine-based organogelators and safflower oil. The outcomes showed that the created organogels were biocompatible and could permanently inhibit the cholinesterase enzyme. Lecithin organogels (LOs) have been thoroughly researched as cutaneous administration vehicles due to their low potential for skin irritation and inherent biocompatibility. Additionally, LOs are very adaptable and compatible with various organic liquids, including different types of food oils, apolar solvents, and medicinal compounds [134].

An ester of sorbitan with palmitic acid is known as sorbitan monopalmitate (SMP). SMP is a hydrophobic nonionic surfactant utilized as a gelator in producing organogel drug delivery systems. Galactose and rhamnose structural units, as well as glucuronic and galacturic acids, make up sterculia gum. It is employed in many food and pharmaceutical businesses because of its ability to retain water. It is a possible candidate for buccal medication administration due to its bioadehsion nature. It works well as a skin and wound healing agent [135]. PLO gel is a yellow-colored, odourless, opaque gel made from soy lecithin with a quick absorption rate. Due to its distinct physical characteristics, it is frequently employed as a medication delivery medium, including poloxamer 407, a substance that increases viscosity and has surfactant capabilities that make oil-in-water preparations easier. Lecithin, isopropyl palmitate, isopropyl myristate, polyethylene glycol, sorbic acid, and potassium sorbate are typical components of PLO gel (Figure 52) [136].

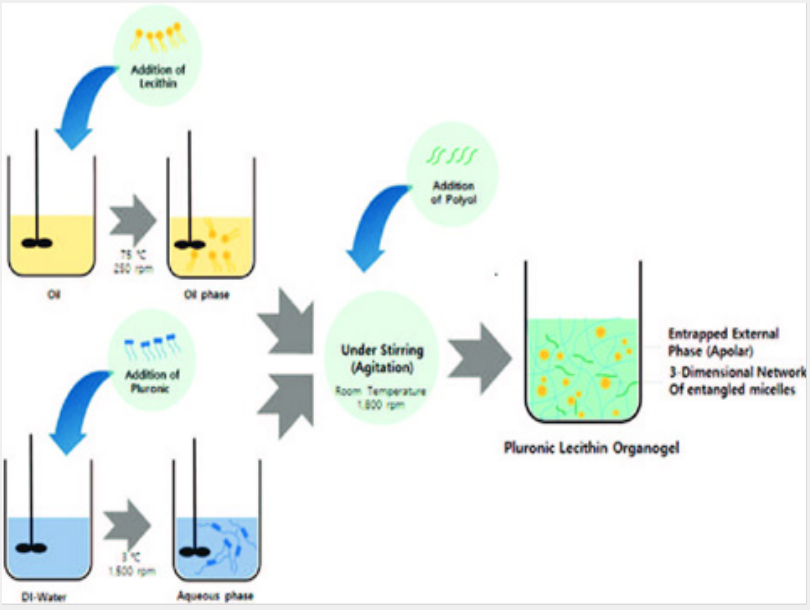

Figure 52: Preparation Method of PLO Gel [136].

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When isopropyl palmitate or isopropyl myristate is used as an apolar solvent, Pluronic lecithin organogel (PLOs), a typical macro-scale supramolecular organogel with high viscosity, is created. Due to their unique design and traits, PLOs can significantly reduce the likelihood of distinct medicinal molecules crystallizing in their micellar structures. A few early examples showed that PLOs were frequently made to load polyunsaturated fatty acids, which have significant therapeutic value for rheumatoid arthritis, cardiovascular disease, and nervous system disorders, and non-steroidal anti-inflammatory drugs (NSAIDs) for treatments of heel pain and piroxicam for the treatment of rheumatoid arthritis.

Drugs can be given through cutaneous, dermal, or transdermal channels into the skin's layers and through percutaneous or transdermal routes outside the skin. These delivery methods prevent first-pass metabolism and produce systemic effects. A molecule's liposolubility determines how bioavailable it is after being applied topically, and this factor might change depending on the type of delivery method used. Due to the stratum corneum's thickness and lipophilic makeup, it serves as the primary barrier. Organogels are a promising class of vehicles with good qualities for cutaneous administration because of their inherent nature. They favour quick absorption and are lipophilic, non-irritating, simple to use, and non-toxic [137].

In Situ Drug Delivery

The market for bio-based polymers is highly active. Self-assembled zein organogels in N-methyl pyrrolidone (NMP), dimethyl sulfoxide (DMSO), and glycerol formal (GF) have been reported by Ali Raza et al. The polarity of the solvent and Hansen Solubility's hydrogen bonding component caused the gel to develop. Gels exhibited thixotropic and shear-thinning characteristics. Additionally, solvent exchange-based in situ implant creation using water-induced zein - a water-insoluble plant protein generated from maize that has various uses in the food and packaging sectors because it may keep prepared goods' humidity levels stable - self-assembly enables mechanically strong in situ implants. As a model drug, ciprofloxacin was added, and sustained release based on the solvent exchange rate was noticed. Over more than 14 days, in situ, implants in agarose gel maintained antibacterial effectiveness against S. aureus (Figure 53). Further applications of zein-based organogels as 3D printing ink revealed that zein gel prepared in DMSO had better printability than gels prepared in NMP and GF [138].

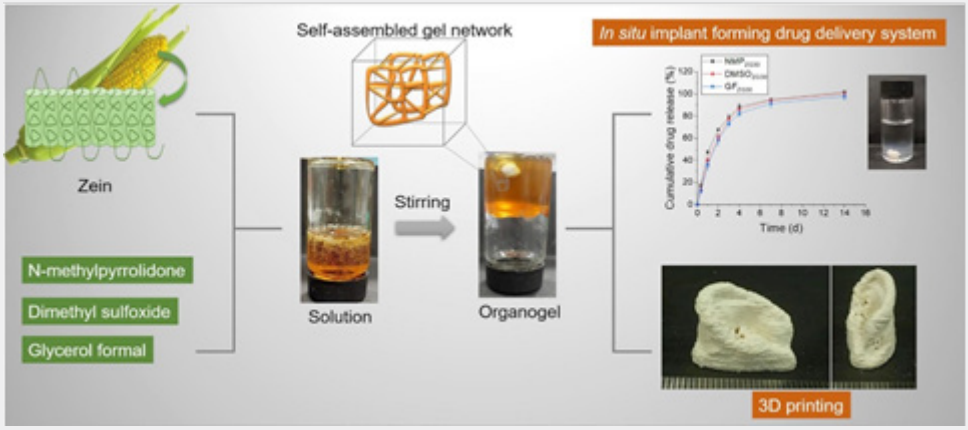

Figure 53: In situ implant forming drug delivery system [138].

It is possible to encapsulate a variety of pharmaceuticals using three solvents when creating organogels, and it is also simpler to create composite gels using additional biocompatible polymers. These organogel systems can also create scaffolds for tissue engineering or 3D-printed medication delivery systems [96].

Organogels integrated parenteral administration (i.e., intravenous or subcutaneous route), which avoids the drawbacks of first-pass metabolism and the harsh environment within the gastrointestinal tract, has received growing interest in drug delivery systems. In situ forming organogel delivery devices have also just recently been created. In situ forming implants for the regulated distribution of hydrophilic acyclovir (ACV) and lipophilic clotrimazole (CTM) were, for instance, demonstrated using 12-HSA-based organogels [139].

Oral Drug Delivery

The use of oral formulations based on organogels to increase the bioavailability of medicinal compounds has been proven effective in the case of an organogel-based nano-emulsion for the oral delivery of curcumin. Based on in vitro bioaccessibility results from lipolysis investigations, tween 20 was chosen as the emulsifier. It was proposed that digestion and diffusion may be the main permeation route for curcumin using the Caco-2 cell monolayer model (Figure 54). The bioavailability of curcumin in the nano-emulsion significantly outperformed unformulated

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curcumin, according to in vivo pharmacokinetic study [140]. To increase the bioavailability of the hydrophilic and lipophilic active substances, organogels may be combined to create oral formulations. Iwanaga et al. studied hydrophilic and lipophilic drug-containing 12-HSA organogels [141]. The use of organogels for additional drug delivery methods, such as the nasal and ocular systems, has been studied. For instance, Dai et al. developed an in-situ organogel to administer poorly water-soluble flunarizine hydrochloride (FNZ) for the treatment of brain illnesses via intraocular administration utilizing soybean oil, stearic acid, and N-methyl-2-pyrrolidinone [142].

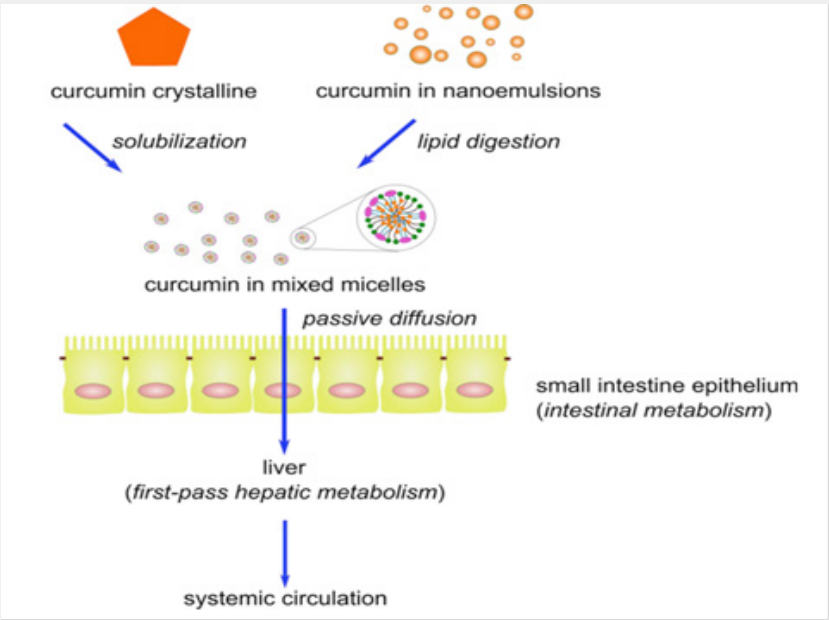

Figure 54: The absorption process of unformulated curcumin and curcumin nanoemulsions [140].

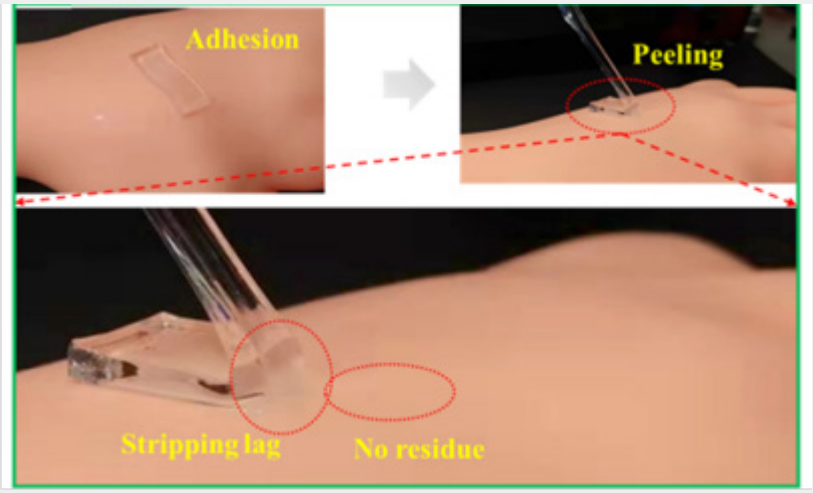

Figure 55: Exhibition of no-residue behavior during peeling from the hand model [143].

Ionic Skin

Organogels are bicontinuous gelatinous materials with intriguing qualities like adaptable elasticity, dynamic adhesiveness, self-healing, and stretchability. They have shown promise in various applications, from flexible electronics and biomedicine to intelligent drive systems. For instance, employing cationic monomers and useful catechol molecules, a polysiloxane-supported organogel elastomer with exceptional elasticity and

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unheard-of adhesive and self-healing behaviours was created. It has been shown that strain-insensitive devices made from conductive polymers and an organogel matrix are electrically conductive, mechanically soft, and seldom dry in ambient air. By combining an amorphous polymer with an organic ionic liquid that is chemically compatible, an artificial skin for aquatic environments was created. It showed transparency, stretchability, and self-healing abilities - all attributed to highly reversible ion-dipole interactions.

By sandwiching a soft organogel between two ionic conductive hydrogels, Zhixing Zhang et al. created a robust capacitive ionic sensor with excellent transparency, mechanical adaptability, and self-healing abilities. The resulting capacitive ionic sensor, which functions as a bioinspired ionic skin, exhibits great pressure sensitivity (0.293 kPa-1), making it simple to track diverse human actions, including finger stretching, wrist bending, and neck movement during chewing. Interestingly, the resulting capacitive sensor can also be fake skin on a pneumatic soft hand, enabling a sophisticated haptic experience (Figure 55) [143].

Bigels

Bigels are biphasic systems made up of two gelled phases: either an organogel dispersed into a hydrogel (O/W), a hydrogel dispersed into an organogel (W/O), or a bi-continuous system (W/O/W/O). Combining both phases in a colloidal system may be advantageous depending on the application because of the synergistic interaction of the organogel and hydrogel. The bi-gels and emulsions produced oil-in-water (O/W) dispersions that contained crystals at the interface between the phases. According to rheological analysis, all samples displayed viscoelastic weak gel behaviour with stronger physical gel interactions when 12HSA was used as an organogelator. Except for sensory measures like thixotropy, shear-thinning, and consistency, the kind of polymer utilized in the hydrogel appeared to disguise the influence of organogels over bi-gels in rheological properties. All samples demonstrated excellent thermal and centrifuge-stress stability, suggesting that additional research may use the 5/95 organogel/hydrogel ratio to compare bi-gels and emulsions. Renata Miliani Martinez et al. provided one viewpoint on the relationship between the rheological characteristics of bi-gels and the particle size distribution. However, more precise models are still required to predict the characteristics of these systems. The usage of bi-gels over the emulsions used to compare in this study would be supported by additional research on the topical distribution of vitamin E from these formulations in ex vivo models. They support the use of stressors to increase sample differences, such as radiation or pollution [144].

Baljit Singh and Rajender Kumar used radiation to create bi-gel based on the hydrogels sterculia gum, poly (AAm), and organogel (olive oil and sorbitan monopalmitate). Their study was unusual because they combined green design principles and green manufacturing techniques to create a bi-gel formulation that served as a medication carrier. They claimed that it falls under the green standard since no additives are used in the radiation procedure to create the gel, and it is a pure and clean process. However, the materials used to make bi-gels are regarded as following green protocol. For existing drug carrier systems, sterculia gum, sorbitan monopalmitate, and olive oil-based bi-gels have been suggested as green substitutes. Triolein (oleic acid, oleic acid, oleic acid) is a liquid lipid in olive oil used to make organogels and bi-gels for topical medication delivery. The possibility for using the moxifloxacin-loaded [olive oil-SMP-sterculia-cl-poly (AAm)] bi-gels as drug delivery systems for peptic ulcer and GIT tract issues is increased [135].

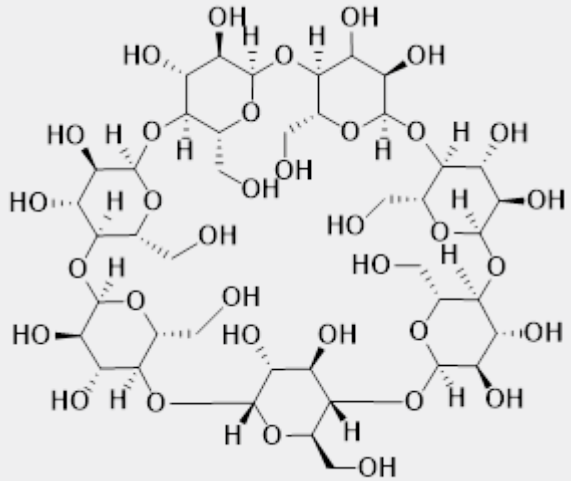

Figure 56: β-Cyclodextrin.

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Anti-Cancer Drug

In recent years, β-cyclodextrin-based supramolecular gels (β-CDs) (Figure 56) have attracted much interest. Conical β-CD gelators with hydrophilic surfaces enable β-CD molecules to self-assemble in cage- or channel-type structures to build soft materials in suitable polar solvents. CD-CD H-bonds, host-guest interactions, and other non-covalent interactions are the primary catalysts for gel formation. To determine whether using medication gels as a method to improve the specific antitumor effectiveness of the chemotherapy drugs 5-Fu or MTX, the cytotoxicity of these drug gels and medications that target human hepatocellular carcinoma cells (HepG2 cells) in vitro was studied by Zhaulou Li et al. They looked at the plain -CD/glycerol gel as a flexible drug carrier. They successfully created this inexpensive medication carrier with excellent business potential for the pharmaceutical sector. Significantly, the drug carrier's extended drug release can help medications to enhance therapeutic effects. Additionally, the mechanism of gel formation depends on CD-CD hydrogen bonding to build -CD fibres and on the crucial hydrogen-bonded network of glycerol to seize -CD gelators similar to mat-rolling. To participate in the gel formation process, the glycerol hydrogen-bonded network is considered as a deciding element [145].

Conclusion

Several of the organic and inorganic compounds are reported to possess gelating ability in different organic solvents. In this mini-review article, special emphasis is given on biomolecule based gelators viz., carbohydrates and amino acids. These organogels are widely used in different areas of science, such as food industry, pharmaceutical industry, lubricant, sensors, cosmetics.

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