

Article

Organogels for Low-Polar Organic Solvents: Potential Applications on Cultural Heritage Materials

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Abstract: Traditional cleaning methods for removing undesired substances from artworks often involve the use of toxic volatile solvents, raising concerns about human health and environmental impact. Over time, various cleaning systems, such as thickeners, rigid, peelable, and nanostructured gels, have been introduced in the conservation sector to minimize solvent use and toxicity. However, these methods are primarily tailored for aqueous solutions or medium-to-high-polar solvents, leaving sustainable organogels for low-polar solvents largely unexplored. This paper explores the application of Low-Molecular-Weight Gelators (LMWGs) in the field of cultural heritage conservation, focusing on their potential to gel low-polar organic solvents. LMWGs, including cholesterol derivatives, fatty acid-derived compounds, anthryl, anthraquinone, amino acid, and saccharide-based organogelators, offer biocompatible and cost-effective options by forming supramolecular gels that immobilize solvents and reduce their release into the environment. This study highlights the need to transition from traditional, often toxic, solvents to greener and more sustainable cleaning systems by emphasizing LMWGs' biodegradability, biocompatibility, and sustainability. While challenges such as optimizing gel properties and ensuring compatibility with artwork surfaces still need to be addressed, LMWGs hold promise as organogelators in conservation practice. Further research into LMWGs should focus on their optimization for conservation applications by adjusting their rheological properties and physico-chemical stability.

Keywords: LMWG; low-polar solvents; non-polar solvents; organogel; green; sustainable; conservation; artworks; cleaning system; cleaning



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1. Introduction

The preservation of cultural heritage materials often demands conservation treatments aimed at removing undesired materials from an artwork's surface. These materials can range from basic dirt to synthetic coatings, intentionally applied by the artist or conservators to protect the object against aging [1–7]. Protective coatings tend to deteriorate over time due to physicochemical reactions taking place in the material. This deterioration results in aesthetic alteration and embrittlement and, consequently, exposes the artwork's surface to dangerous external agents [8,9].

The “cleaning” intervention is aimed at restoring the object's aesthetic value by removing the degraded coating or soiling deposit, often involving the application of a new coating capable of effectively preserving the remaining original materials. This task can be delicate for conservators, as they must operate without damaging the layers that need to be

preserved. This involves avoiding mechanical stress, chromatic alteration, and the removal of the original components [10].

Recent advances in the study of greener formulations have introduced new sustainable solutions. However, despite the well-known hazards to human health and the environment posed by solvents like methylethylketone, aliphatic, and aromatic hydrocarbons, these substances continue to be widely used in common restoration practices [11–18]. This conservative approach can be attributed to conservators' initial skepticism towards innovative greener solutions. New products often differ in terms of effectiveness, application methods, and duration compared to the familiar ones, making conservators hesitant to adopt them. Additionally, these innovative solutions require rigorous testing and validation by professionals before they can be safely applied to cultural heritage materials. Moreover, the dissemination of testing and validation results to end-users is often insufficient, further hindering the adoption of these alternatives. Even after conducting testing and validation procedures, their findings are rarely adequately communicated to the end users.

For a considerable period in the field of restoration and conservation science, the primary focus was on finding solutions that were highly compatible with the materials of artifacts, occasionally overlooking potential concerns with regard to both the operators and the environment [19]. Indeed, the low cost, transparency, and purity of traditional solvents are highly desirable features for conservators, along with their medium-to-high volatility, which prevents the retention of the solvents in the artwork's substrate. However, the interaction between the free solvent and the artwork's materials is not entirely controllable and might lead to either the diffusion of the solvent in the substrate's porous matrix or the swelling or partial solubilization of the organic components, if present. Eventually, the diffusion of the solvent in the substrate might also cause the transportation of the materials to be removed within the porous matrix. This can induce the further darkening and deposition of undesired materials in the artwork's substrate. Hence, the selectivity of the cleaning system is of the utmost importance and serves as a preventive measure to avoid further damage caused by the conservation treatment [11].

Within this context, the integration of materials science, colloid science, and interface science paradigms into conservation practices has brought about a revolutionary shift. This has, in turn, led to significant advancements in the creation of environmentally friendly functional materials for the cleaning of cultural heritage over the years. Cleaning systems such as thickeners (cellulose ethers, polyacrylic acid-based, and modified silicone-based formulations), poulticing materials (soft fiber mats and synthetic clay), and rigid gels (agarose and gellan) have been introduced into the conservation sector to reduce the volatility of the solvent and limit its diffusion on the artwork's surface and in the environment (Figure 1) [20–22]. These systems have been commonly used in the conservation sector based on the specific requirements of the cleaning treatment. Thickeners form flexible viscous gels that can adapt to the surface and be kneaded with brushes or cotton swabs to further enhance removability. However, the direct application of these gels onto the artwork may result in the release of gel's residues on the surface. Typically, a "clearing agent", which is the same solvent entrapped within the gel, or an intermediate layer of Japanese paper is used to mitigate the risk of leaving residues on the artwork. To address this issue, rigid physical gels made of polysaccharides, such as agarose and gellan, have been introduced to prevent the "residue" problem and facilitate the easy removal of the gel after the cleaning treatment. However, these gels face challenges in conforming to embossed or 3D surfaces, and the residue problem has not been completely resolved. Ongoing research aims to specify the pore size of these gels and control solvent release [21,23].

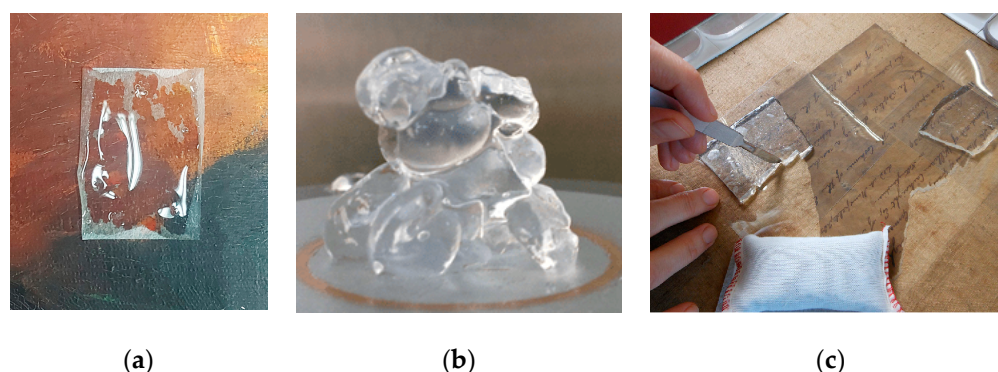


Figure 1. Examples of cleaning systems used in cultural heritage conservation: (a) 5 wt% of hydroxypropyl cellulose (Klucel® G) in a water-based solution applied on an oil painting by interposing a sheet of Japanese paper to limit the release of residues on the surface; (b) 3 wt% of synthetic clay (Laponite®) in H₂O (image taken from “Experimental Study of Complex Fluids”) [24]; (c) water-based solution gelled with gellan gum used for the removal of a paper label from the verso of an oil painting (image taken from Maheux 2015) [25].

A significant improvement was represented by the development of advanced colloidal systems, such as peelable and nanostructured chemical gels (Figure 2) [20,26,27]. These innovative tools are created through a crosslinking reaction between polymers, yielding more stable products with known mesh sizes and release rates and the ability to adapt to surfaces with different morphologies [21]. Among their advantages, they require small amounts of solvent, leading to the controlled release of the continuous phase on the artwork’s surface and a consistent reduction in toxicity and environmental impact. In the last decade, considerable effort has been devoted to the development of nanostructured peelable gels, including acrylamide gels, polyhydroxybutyrate-based gels (PHB), polyvinyl alcohol-based gels (PVA), and semi-interpenetrating polymer networks of poly(2-hydroxyethyl methacrylate) [p(HEMA)] and polyvinylpyrrolidone (PVP) [18,26,28,29]. The low toxicity of these formulations is primarily due to the immobilization of the cleaning agent within the 3D polymer network. Entrapping the solvent results in the controlled and reduced release of solvent on the artwork, which limits both the amount of solvent used and its inhalation by the operator. Additionally, the cohesive forces of these gels surpass the adhesion forces on the substrate, facilitating the removal of the gel from the artwork’s surface without leaving residues.

For instance, peelable polyvinyl acetate-borax polymeric dispersions were utilized to remove the carbonaceous particles deposited on the painted surface of the *Giudizio Universale*, a fresco by Carlo Carrà (1881–1966), to prevent the swelling or damage of water-sensitive materials, such as gypsum and white egg [30]. Similarly, semi-interpenetrating polymer networks of p(HEMA) and PVP were employed to remove grime from the surface of Thang-Ka, which is a *Tempera magra* painting on canvas typically featuring a very low-cohesive painted layer with a small amount of a water-sensitive binder (Figure 2b) [29,31].

Both traditional and emerging cleaning systems mentioned above primarily involve hydrogels or organogels designed to retain aqueous solutions or medium-to-high-polar solvents. So far, only a few solutions are available in the cultural heritage conservation sector for the thickening of low-polar and non-polar solutions. Nevertheless, these solvents are commonly used in conservation practices. Aliphatic hydrocarbons such as Petroleum Ether and Mineral Spirits are less likely to react with acids, bases, or other reagents due to the presence of inert C–C covalent single bonds in their structures. Consequently, they can be more safely applied on various materials and only interact with other non-polar compounds. For this reason, they are commonly used to remove natural and microcrystalline waxes from artworks, such as outdoor stone and metal sculptures [5–7,15,32–34]. Simultaneously, the removal of natural and synthetic varnishes, both aged and unaged, requires solvents with different degrees of polarity based on the nature and conservation state of the coating.

Low-polar solvents are often mixed with polar compounds to achieve the desired degree of polarity, depending on the substance that needs to be removed [11,35,36].

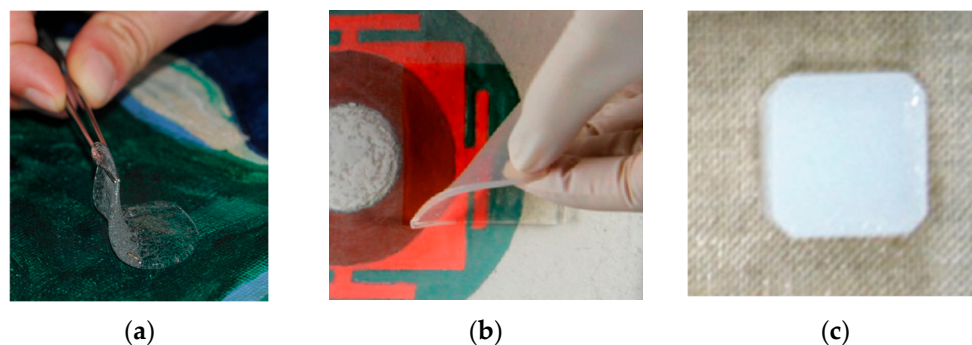


Figure 2. Examples of cleaning systems used in cultural heritage conservation (images taken from Baglioni et al. (2013) [31]: (a) peeling of a PVA/borate via highly viscous polymeric dispersion from a paint surface; (b) semi-interpenetrated network of PVP and pHEMA used for the cleaning of a water-sensitive paint; (c) oil-in-water (O/W) microemulsion confined inside an acrylamide gel.

Despite the widespread use of low-polar solvents in the conservation sector, research into potential organogelators capable of reducing the impacts of these solvents is still in its early stages. Currently available solutions mainly consist of thickeners and silicone-based gels [20,21,26,27,37,38]. These gelators have different drawbacks, ranging from the toxicity of the gels' constituents to the release of residues onto the artwork's surface [38]. Recently, sustainable organogels based on castor oil, poly(hexamethylene diisocyanate) (PDI), and polyethylene glycol (PEG) were proposed to gel *p*-xylene and *n*-pentanol for the removal of aged acrylic–ketonic varnishes [39]. However, effective greener gelators for low-polar and non-polar solvents are still lacking.

Potential sustainable candidates could be drawn from the drug-delivery sector. Indeed, biocompatibility, biodegradability, and no toxicity are essential qualities for the use of hydrogels or organogels in the biomedical field, where subcutaneous injection or oral drug delivery applications are common [40]. Delivery systems have been extensively studied in the last few decades to achieve the desired therapeutic effect by controlling and targeting drug release. The porosity of hydrogels and organogels porosity allows the loading of drugs into their 3D network, releasing them at a rate dependent on the molecule's diffusion coefficient through the gel network. This controlled release ensures that the drug reaches the target site and avoids issues associated with the drug's rapid release in the human body [40,41].

Among these delivery systems, Low-Molecular-Weight Gelators (LMWGs) have found widespread use not only in the pharmaceutical industry but also in the food and cosmetic sectors over the last three decades. These naturally occurring, cost-effective molecules can encapsulate both hydrophilic and lipophilic constituents through a straightforward preparation process [42]. LMWGs have molecular masses lower than or equal to 3000 and have demonstrated the ability to gel water or organic solvents, even in small amounts of 2 wt% [43,44]. The resulting gel comprises a supramolecular structure composed of physically self-assembled fibrils linked via non-covalent forces, creating a 3D network that immobilizes the solvent [45,46].

Given the need for developing organogels capable of retaining low-polar and non-polar solvents while ensuring their controlled release and biocompatibility, various LMWGs are presented herein to assess their suitability for application in cultural heritage conservation.

2. Gels for Low-Polar Systems in Cultural Heritage Conservation

The main thickeners/gels available in cultural heritage conservation are presented hereafter to provide insights into the materials currently utilized in this sector to thicken low-polar and non-polar organic solvents.

2.1. Chemical Solvent–Surfactant Gels

Solvent gels are complex systems comprising a surfactant and a polymer, indirectly thickening the solvent in a small aqueous phase, which is mixed or emulsified into the solvent phase [38]. Typically, a solvent gel is made of a polyacrylic acid (PAA) and an alkaline surfactant, often a polyoxyethylene tallow amine, along with water and an organic solvent. Carbopol® (PAA) has been widely used in the cultural heritage sector to reduce solvent volatility. It is combined with tertiary amine ethoxylate surfactants, such as Ethomeen® C/12 or C/25, derived from primary coco amines. These solvents are used to deprotonate the PAA, forming amine carboxylate salts, thus creating a salt (ionic) link with the carboxylic groups on the Carbopol®. During gel formation, Carbopol®'s polymer chains unfold in the solution. Solvent gels' structures address two critical issues in artwork cleaning: the entrapment of the solvent in the polymeric network limits its diffusion into the object's porous matrix, while the surfactant (Ethomeen®) acts through its detergency ability. With different hydrophilic–lipophilic balance (HLB) values, C/12 and C/25 Ethomeen® can be used to thicken liquids with varying degrees of polarity, ranging from alcohols and ketones to aliphatic and aromatic hydrocarbons. These polymer dispersions have proved to be effective and versatile, remaining a standard tool for the cleaning of works of art, primarily employed for varnish or adhesive removal from different types of artifacts [38,47]. However, previous research has indicated the presence of surfactant residues on the surface. Ethomeen® was identified as the major component of the gel residue compared to Carbopol® [38,48–50].

2.2. Silicone-Based Thickeners

A recent update by Wolbers introduces the use of modified silicone thickeners based on cyclopentasiloxane and dimethicone cross-polymers, such as Velvessil Plus™ [21]. These dispersions are considered less risky on water-sensitive surfaces compared to free solvents or aqueous solutions, which may cause the extraction of additives or surfactants originally included in the paint layer.

Gelling agents based on silicone polymers incorporate ethoxylate/propoxylate chains or polyether chains to bridge the gap between silicone chains. The polar groups can emulsify water into the silicone system, while the silicone side chains are compatible with silicone solvents, and hydrocarbon side chains are compatible with many other conventional solvents, such as Petroleum Ether or Ligroin [51]. Emulsions made with these polymers can hold acetone, acetone/alcohol mixes (i.e., acetone/isopropanol), or benzyl alcohol [52]. Specifically, Velvessil Plus™ gels with benzyl alcohol and Velvessil Plus™ gel with acetone have been employed to assess the removal of permanent marker ink from historical parchment, while Velvessil Plus™ coupled with Cyclomethicone D5 was used to eliminate soiling deposits from varnished brass [53,54]. However, the need for a clearance step to remove polymer residues has limitations for the use of these systems, as the clearing solvents are the same cyclomethicones uploaded in the viscous dispersion. Additionally, aside from the health risks associated with the silicone solvents recently identified as toxic compounds, the currently used solvent (decamethylcyclopentasiloxane, D5) has a relatively high boiling point (210 °C), increasing its permanence in the artifact's layers [55].

2.3. Emulsions and Microemulsions

2.3.1. O/W Emulsions

An emulsion is a stable dispersion of two or more liquids that are not miscible under normal conditions, being held in suspension by small percentages of substances called “emulsifiers”. Emulsions can simultaneously combine the action of two immiscible solvents. For instance, White Spirit and water can be emulsified to remove both polar and non-polar materials from a painted surface, where water acts on the polar substance and White Spirit acts on the non-polar one [36]. Emulsions can be oil-in-water (O/W) or water-in-oil (W/O), depending on the continuous phase (usually, the more abundant one), where “oil” is any hydrophobic substance, including non-water miscible solvents (e.g., aliphatic, aromatic,

hydrocarbons, halogenated materials) [56,57]. A third type of emulsions is known as “inversion-type emulsions”, which tend to combine both “oil” and “water” phases with oil and water phase combinations at nearly equal molecular volumes, i.e., at their phase inversion point [57]. Emulsions are stabilized using surfactants (emulsifying agents) that have affinities for both phases, reducing the interfacial surface tension, i.e., the energy required to produce new surfaces between the two phases. Non-ionic emulsifiers like Triton X-100, Tween 20, and Brij 35, which have a low hydrophile/lipophile balance (HLB), are emulsifiers commonly used in the conservation of cultural heritage [57,58]. These emulsifiers allow the entrapment of the oil phase in the polar (or water) phase or vice versa while aiding in wetting out and picking up dirt [36]. The O/W and W/O arrangements create a gel-like system, where the contact between the dispersed phase and the water-sensitive or solvent-sensitive material is restricted or stunted [57]. Ultimately, the presence of the emulsifier necessitates a clearance step based on the type of emulsifier used, typically involving the use of aliphatic hydrocarbons.

2.3.2. Inorganic Emulsions

Most recently, new nanostructured fluids have been explored, including inorganic and Pickering emulsions, to discover innovative applications. Halloysite nanotubes modified using sodium dodecyl sulfate (SDS) have been suggested for formulating O/W emulsions, proving efficient in removing wax from marble artifacts [59,60]. The unitary cell formula is $\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4 \cdot n\text{H}_2\text{O}$, similar to kaolinite, except for the presence of the water molecules (typically 2) hosted between adjacent clay layers. The surface chemistry of halloysite nanotubes is compelling due to the differences between the inner and outer surfaces: the external surface comprises Si-O-Si bonds, while the inner surface comprises a gibbsite-like array of Al-OH groups. This structure enables targeted modification through selective functionalization and results in charge separation within the nanotube. For instance, composites of sodium alkanoate/halloysite nanotubes can adsorb aliphatic (*n*-decane) and aromatic (toluene) hydrocarbons, trapping them in the hydrophobically modified lumen [59].

In terms of applications, halloysite’s notable features include surface reactivity, a hollow cavity, easy dispersibility and stability in solvent media, and long aspect ratios. An alternative approach to stabilizing oil in aqueous solvent is through Pickering emulsions, which rely on solid particles with exceptional colloidal stability as a consequence of their opposition to coalescence processes and Oswald ripening [61].

3. Organogels Structure

Gels form through hierarchically ordered 3D microstructures that trap solvents, controlling their fluidity. They are classified as physical gels or chemical gels, with physical gels resulting from the self-assembly of low-molecular-weight organic molecules through weak intermolecular interactions, such as hydrogen bonding, electrostatic interactions, van der Waals forces, and π - π interaction. Chemical gels, on the other hand, are covalently crosslinked network polymers that are typically thermally irreversible [62,63]. Physical gels are further categorized into hydrogels and organogels based on the entrapped solvent.

When a solute and a solvent are mixed, various outcomes can occur, including precipitation, dissolution, crystallization, or gelation. Precipitation or crystallization results when solute–solute interactions are stronger than solute–solvent interactions, leading to the formation of solid particles or crystals. Conversely, dissolution occurs when the solute–solvent interaction dominates, causing the solute to dissolve in the solvent. Gelation, on the other hand, happens when there is an optimal balance between the solute–solute and solute–solvent interactions. This balance, often assisted by amphiphilicity, results in the self-assembly of molecules in one dimension, leading to the formation of superstructures with high aspect ratios, such as fibers or ribbons, defining the process of gelation. On the contrary, crystallization results from self-assembly in all dimensions of space with a more or less similar speed of propagation [62].

Terminology for organogels remains varied and lacks standardization. Different definitions co-exist [64–66], but a more comprehensive definition of organogel was proposed by Kuzina et al. (2023) as “colloidal, supramolecular, or polymer 3D networks filled with organic liquids” [67]. The solvent’s role in the gelation process is significant. The solvent’s structure can either initiate or impede stable gelation, highlighting its crucial role in determining the success of the gelation process [68–71].

While hydrophobic forces play a significant role in the aggregation of fibers in water, the gelation in organic media is driven by different interactions. In 1997, Terech and Weiss defined the dipolar interactions and, to some extent, specific intermolecular hydrogen bonds or metal coordination bonds as the main driving forces leading to the aggregation between gelator molecules, thus forming the lamellar or fibrillar structure [72]. Based on the explanation of the structure formation given by Liu in 2005, self-assembled fibrillar networks are formed as 3D structures through initial fiber nucleation, repeating crystalline fiber branching, and fiber growth. The self-assembly process is primarily controlled via a balance between physical non-covalent interactions, such as hydrogen bonding; London dispersion forces; π - π stacking; and electrostatic forces [43,73]. As a consequence, stable and flexible gelation can be achieved in solvents with limited non-covalent interactions with the gelator, as the structure of the final gel, as well as its thermal and environmental stability, is significantly affected by the nature of the solvent [67].

Due to the need for the formation of intermolecular physical interactions, specific functional groups have been found to cause the formation of gels. Specifically, carbohydrates consisting of multiple hydroxyl groups (-OH) and nucleobases tend to easily form hydrogen bonds due to $O\cdots H$ physical bonds and π - π stacking interactions, respectively. Also, long-chain alkanes produce van der Waals forces, while amides (RCONHR') and ureas (RNHCONHR') form hydrogen-bonding interactions due to the $C=O\cdots H-N$ bond [43].

When the gel is formed, it can be either hydrophilic, hydrophobic, or amphiphilic in nature based on the polarity of the gelator and the features of the solvent. It acts like a quasi-solid material whose surface does not behave like a generally solid surface. Indeed, wettability of the substrate by the gel is defined by the liquid surface layer formed via the extrusion of the solvent. In addition, the gel stability is significantly affected by the boiling point value of the solvents. Lower boiling point solvents tend to evaporate quickly from the 3D network, leading to the shrinkage of the gel and affecting its mechanical and thermal stability, while higher boiling point solvents contribute to gel stability [67].

4. Low-Molecular-Weight Organogelators (LMWGs) for Non-Polar and Low-Polar Solvents

Even though some possibilities are already used in the cultural heritage sector for thickening non-polar and low-polar substances, greener alternatives can be found among Low-Molecular-Weight Gelators (LMWGs) that are commonly used in the pharmaceutical industry, as well as in other sectors.

Low-molecular-weight organogelators (LMWGs) have attracted increasing interest as natural materials with applications in drug delivery and tissue engineering. They have been used for a variety of applications so far, such as scaffolds for the mineralization of hydroxyapatite and as matrices for the growth of calcite crystals. Other applications include their use as gel electrolytes, liquid crystals, photochemical usage, pharmaceuticals, and methods of drug and gene delivery [45]. These compounds have molecular masses of ≤ 3000 and can gel water (hydrogels) or organic solvents (organogels) in small amounts (typically ≤ 2 wt%) by creating a 3D continuous phase mainly composed of the liquid and, to a lesser extent, the solid [43,44]. They form supramolecular gels mostly composed of nanofibers, tapes, rods, nanotubes, ribbons, or sheets hierarchically self-assembled as a result of non-covalent forces, such as hydrogen bonding, van der Waals, π - π stacking, coordination, electrostatic and charge-transfer interactions, creating a three-dimensional network where the solvent is immobilized [45,46]. The self-assembly depends on the physical interactions between the gelator’s molecules, which have to form aggregates long

enough to overlap and cause the gelation of the solvent [44]. The mechanism leading to the formation of the gel commonly consists of the dissolution of low percentage quantities of the gelator (0.1–5 wt%) in a heated solvent. Upon cooling, the affinity between the liquid phase and the gelator decreases, leading to the self-assembly of a 3D network of solid fibers, in which the liquid is immobilized (Figure 3) [43,74]. LMWGs can be composed of solid or fluid fibers. Solid-matrix gels have solid-like networks with large junction points [75]. Fluid-matrix gels behave just like solid-matrix gels, except for the kinetic behavior of the matrices. The solid ones show robust morphologies, while the fluid ones form transient structures in constant dynamic remodeling.

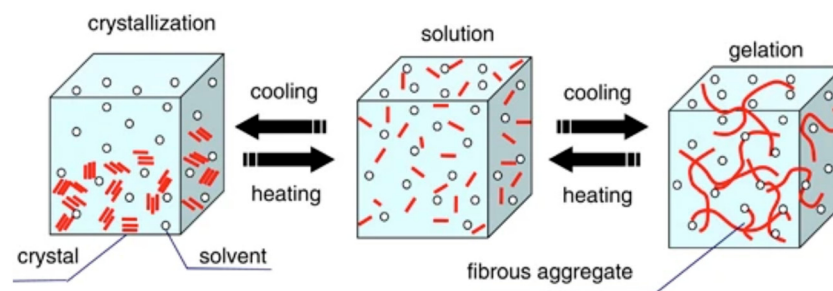


Figure 3. Crystallization and gelation of low-molecular-weight compounds (image taken from Hanabusa et al., 2014) [76].

Many organic compounds can be used as LMWGs, in particular fatty acid derivatives, anthryl derivatives, steroid derivatives, amino acid types, and saccharide and organometallic compounds [42,77].

4.1. Cholesterol Derivatives

Cholesterol has been used as a starting material for the synthesis of steroid-based molecules due to the easy derivatization of its hydroxyl groups, its availability, and its low cost [78,79]. It is considered a rigid and almost planar lipid-type molecule that comprises four essential domains (I–IV) (Figure 4). The polarity of the 3-hydroxy group (I) represents an active site for hydrogen bond interactions, while the absence of methyl groups at C-4 and C-14 (II) is one of the main causes of the molecule's planarity. The right-handed conformation of the side chain is caused by the natural (*R*) configuration at C-20 (III) and particularly significant for intermolecular contacts (IV). Also, the 3-hydroxy group on the A-ring, together with a hydrophobic hydrocarbon body, provides a molecule with an amphiphilic nature [80]. In these organogelators, hydrogen bonding, van der Waals interactions, and π - π stacking are the main forces guiding the formation of long fibers and helices, with this process being governed by crystalline growth through nucleation and the results in the 3D network [42].

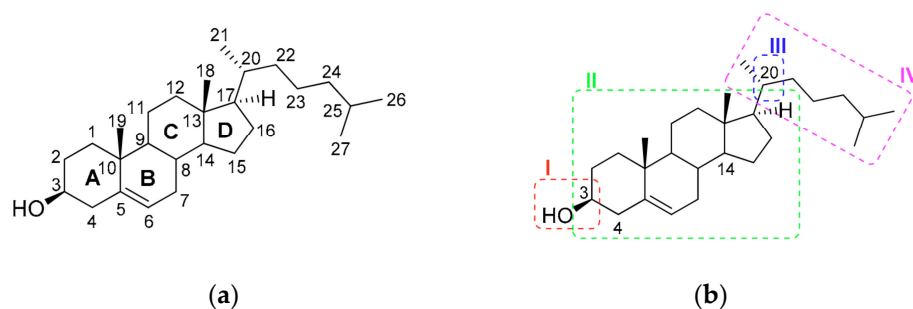


Figure 4. (a) Numbering and ring-labeling convention in cholesterol; (b) four domains of cholesterol [80].

Different polymerization methods and chemical reactions allow the synthesis of polymers bonded to cholesterol in either the side chain or the main chain. Based on the method-

ology of their preparation, polymers comprising cholesterol can be divided into four categories. They can be obtained either via the polymerization of cholesterol-containing monomers, via the post-modification of sidechains, using a chain transfer agent or an initiator containing cholesterol, or as a result of a chain-end post-modification chain [80,81].

Weiss, Fang, and Shinkai studied ALS-type compounds, based on an aromatic moiety (A), which through a linker group (L) is bound to a steroidal group (S) [82]; dimeric cholesteryl derivatives [83]; and pyridine cholesterol-based compounds [84], respectively. In addition, bile acid-based organogelators have been reported in [85].

Among sterol organogelators, ALS can immobilize a wide range of polar (protic and aprotic) and non-polar solvents according to the features of the three components [42]. Solubility is influenced by the chemical nature of the aromatic group, chirality, and the length and flexibility of the linker group.

Other types of gelators comprise organometallic cholesterol-based organogelators, which might consist of derivatives of ferrocene, titanocene, copper, zinc, rhodium, iridium, and palladium, which can add new functionalities due to their variable oxidation states, coordination geometries, and the co-ligand sphere offered by the metal ions [86–93]. Amino-acid sterol-based gelators, such as cholesterol or bile acids with steroidal backbones, have been also found to form organogels. While some of them can only cause the gelation of aromatic solvents, diacid monoamides of cholesteryl glycinate ammonium carbamates were found to be ambidextrous gels able to gelate both non-polar solvents and water [94–96].

4.2. Fatty Acid-Derived Organogels

The carbon chain length of fatty acids defines their gelling ability [97]. They form hydrophobic interactions between the chains and hydrophilic forces between the acid moieties, resulting in a 3D network that encloses organic solvents.

Sorbitan monostearate (Span 60) and sorbitan monopalmitate (Span 40) organogels were first reported by Murden et al. as agents able to gelate organic solvents in both the presence and absence of an aqueous phase (Figure 5) [98–101]. They were able to obtain anhydrous thermoreversible gels by dissolving 1–10% of the organogel in C5 alkanes, isopropyl myristate, and other vegetable oils at 60 °C and letting them cool. The eventual addition of water upon cooling led to the formation of W/O or vesicle-in-water-in-oil (V/W/O) organogel systems. Even though Span 60 is the most extensively studied fatty acid-derived organogel, other sorbitan monoesters, such as Span 40, and 12-hydroxystearic acid have been used for various vegetable oils and hydrocarbons [102,103].

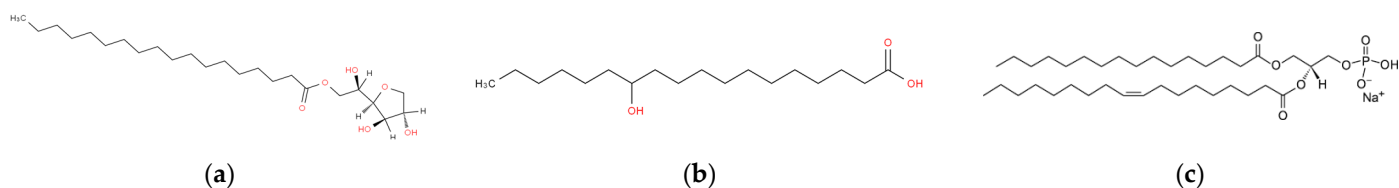


Figure 5. Structures of some fatty acid-derived organogelators: (a) Span 60; (b) 12-hydroxystearic acid; (c) lecithin.

Among the properties affecting organogel formation, chirality also plays an important role, since chiral centers are favorable to fiber growth [104]. 12-hydroxystearic acid can gelate mineral oils by only forming cyclic dimers at a concentration <1.0 wt% in its enantiopure D-12-hydroxystearic acid form, while the racemic mixture could not gelate the oil even at 2.0 wt% due to the formation of acyclic dimers [105]. Mallia et al. studied the structure of (R)-12-hydroxystearic acid (HSA) and demonstrated that H bonding between amides can be stronger than between amines and the N-alkyl group (Figure 5). Also, charged centers at the head groups (NHCO_2^-) can modify the molecular packing of the gelator. Data showed that the introduction of the hydroxyl group to stearic acid (SA) and the presence of an amide group increased the efficiency of the gelator [103].

Lecithin (phosphatidylcholine) is a natural, biocompatible amphiphilic phospholipid derived from soybeans and egg yolk containing fatty acid fractions (Figure 5). Its amphiphilic structure makes it form organogels when mixed with organic solvents due to the formation of isotropic reverse tubular–micelle solutions, while it produces a sort of liquid crystalline order in the case of binary water–lecithin systems [106]. The micelles immobilize the continuous phase (non-polar organic solvent), resulting in the formation of a 3D network. The spherical reverse micellar state of the lipid aggregates opens up, forming tubular micelles when water is added, thus becoming entangled to form a 3D network in the solution bulk [107].

Past studies have shown that hydrogenated soybean lecithin and synthetic lecithin containing residues of saturated fatty acids could not form organogels, proving the importance of naturally occurring lecithin for gelation [107–109]. Linear, branched, and cyclic alkanes, as well as ethers, esters, amines, and fatty acids, can be gelled in combination with a polar agent, i.e., water, which acts as a structure-forming and stabilizing agent [110].

4.3. Anthryl- and Anthraquinone-Derived Organogelators

Common anthryl and anthraquinone organogelators include 2,3-didodecylxytetracene (DDOA) and 2,3-dihexadecylxytetracene (DHDOT), which are able to immobilize organic solvents, such as alkanes, alcohols, cyclohexane, halogenated molecules, etc., due to the presence of the anthracene moiety that allows the formation of dipolar interactions, van der Waals forces, and π - π stacking ability [72,101,111,112].

4.4. Amino Acid-Type Organogelators

L-amino acids are naturally present in all living bodies; thus, they are biodegradable, biocompatible, non-toxic, and eco-friendly. Amino acids such as L-glycine, L-alanine, L-valine, L-isoleucine, L-leucine, L-lysine, L-phenylalanine, L-tryptophane, L-serine, and L-tyrosine are included in the LMWGs class since they are zwitterionic, have polar and non-polar subunits, and have long alkyl chains with an amino group and aromatic ring [42,45]. The presence of the positively charged NH_3^+ group and, sometimes, the aromatic ring cause the gelation due to the formation of hydrogen bonds and π - π interactions, respectively. For these reasons, they act like biosurfactants, as they have different gelation properties based on the type and number of amino acid side groups [113,114]. Non-polar solvents have found to be gelled by the phenylamine derivatives of N-protected phthaloylhydrazide amino acids and a two-component organogelation system based on N-dodecyl-L-amino-acids and N- ϵ -dodecyl-L-lysine esters, which act as the acidic and the basic components of the gelling system, respectively (Figure 6) [113,115,116]. Also, alanine derivatives, N-lauroyl-L-alanine, N-lauroyl-L-alanine methyl ester, and N-steroyl-L-alanine (m)ethyl esters are able to form organogels with vegetable oils, while hydrocarbons are physically gelled by fatty acid amides derived from natural amino acids [117–122]. Furthermore, Palet et al. showed that the fatty acid amides of natural amino acids can gel at room temperature in different organic solvents [123]. They also reported that n-hexane and n-heptane gelation were dependent on the lipophilicity of the fatty acid amides of L-amino acids molecules and occurred with alanine derivatives with chain lengths of C14 or higher [123].

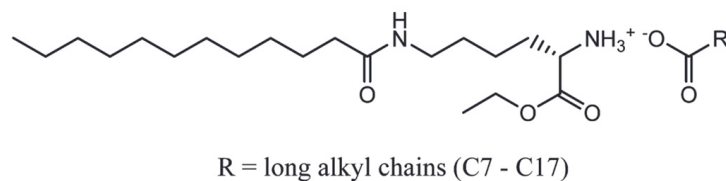


Figure 6. Two-component organogelation system consisting of L-lysine derivatives and aliphatic acids with long alkyl chains [115].

4.5. Saccharide-Based Organogelators

Sugar-based organogels can be identified based on the presence of α -glucose and an aromatic ring in their structure [124,125]. The latter interacts with the non-polar solvent, while the 3D structure is created via hydrogen bonding between the sugar moieties of the gelator. Carbohydrates are, thus, also able to self-assemble in non-polar solvents by forming hydrophilic building blocks due to the presence of multiple hydroxyl groups able to favor solubilization in water [46]. The molecular structure of the gelator strongly affects the stability and gel properties. Indeed, while the main interactions directing the formation of supramolecular gels are given by the presence of -OH groups, the presence of an aromatic group or a hydrophobic moiety favors π - π stacking or van der Waals interactions, respectively [46].

Carbohydrates offer multiple prospects for functionalization. Prathapet et al. reported the partial protection of hydroxyl groups of mannitol as ketals yielding amphiphilic diols, which proved to be efficient gelators for various non-polar solvents and oils [62]. Indeed, the vicinal diol functionality and ketal-protecting groups seem to represent important structural features (Figure 7) [126]. Soundharajan et al. also reported the synthesis of 4-O-octyl substituted oxadiazole N-glycosylamine that can form stable gels in both polar and non-polar solvents [127]. The multifunctional low-molecular-weight organogelators derived from mannose with rational protecting group manipulations were found to give excellent gelation with different organic solvents, oils, and petrol via sequential heating-cooling treatment, providing either transparent or opaque gel at ambient temperature [128].

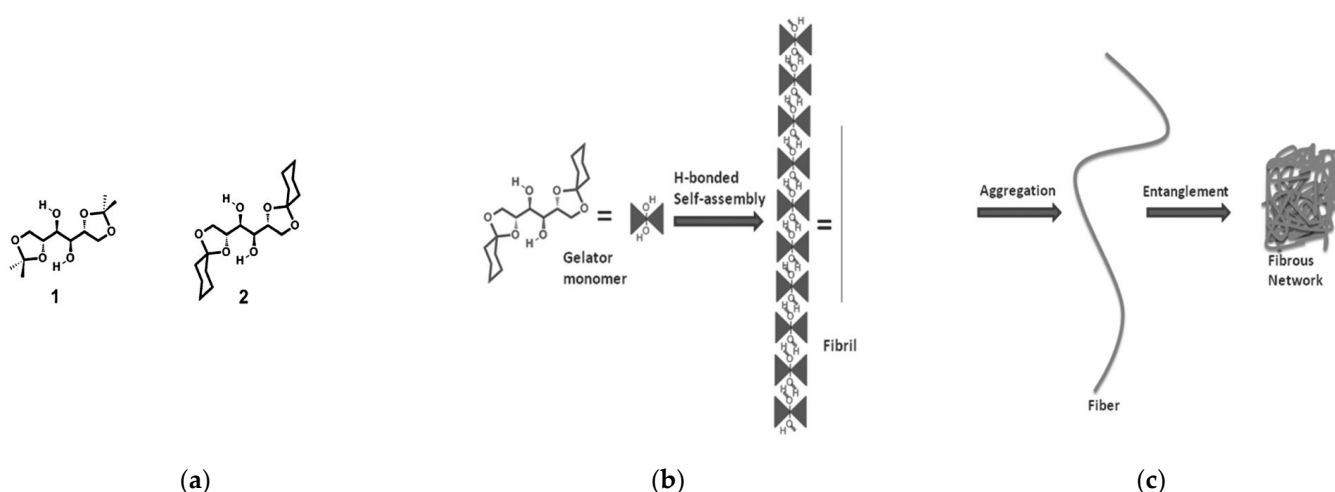


Figure 7. Mechanism of gelation using diols 1 and 2 in non-polar solvents: (a) mannitol-based diols 1 and 2; (b) H-bonded self-assembly; (c) fiber aggregation and entanglement [126].

5. Discussion

The gelation ability of LMWGs is significantly influenced by the nature of both the solvent and LMWG due to specific non-covalent interactions between the two compounds. Organogelators like sterol-, anthryl-, anthraquinone-, and saccharide-based compounds might have limited ability to gel linear aliphatic hydrocarbons and oil-based solvents with linear alkyl chains. The formation of the supramolecular structure in these cases relies on the presence of -OH groups, aromatic groups, and cyclic hydrophobic moieties. These functional groups favor interactions with water or alcohols (through hydrogen bonds), aromatic hydrocarbons (via π - π stacking interactions), and cyclic hydrocarbons (involving van der Waals interactions and London dispersion forces, enhanced based on structural similarities with the hydrophobic moieties), respectively, rather than linear aliphatic hydrocarbons. Indeed, cholesterol- and saccharide-based compounds are more likely to form gels in cyclic aliphatic hydrocarbons compatible with their hydrophobic regions. The presence of cyclic structures in these compounds, such as glucose or galactose rings in

the saccharide, may hinder their compatibility with certain linear aliphatic hydrocarbons, potentially limiting the gelation process. In addition, the gelation ability can be further hampered if the hydrophobic–hydrophilic balance of the gelator molecule is not suitable for the solvent in question [129]. However, the substitution of a functional group with a long aliphatic alkyl chain would favor van der Waals interactions with linear aliphatic hydrocarbons and oil-based solvents with linear alkyl chains [46,130].

In the case of amino acid type gelators, the presence of an aromatic ring and positively charged -NH_3^+ groups often favors the formation of π – π stacking interactions and hydrogen bonds, respectively. At the same time, the presence of long alkyl chains promotes the interaction with linear aliphatic non-polar solvents due to London dispersion forces occurring between linear alkyl chains. Indeed, amino acid derivatives with long hydrocarbon chains, such as N-lauroyl-L-alanine, N-lauroyl-L-alanine methylesters, and N-steoyl-L-alanine (m)ethyl esters, were found to self-assemble in situ in the presence of vegetable oils due to structural similarities between the hydrophobic moieties of the gelator and the linear alkyl chains of the solvent [42].

In general, the presence of a substituent with a long linear alkyl chain tends to improve the ability of the gelator to form a supramolecular gel with linear aliphatic hydrocarbons. The linear chains overlap, allowing optimal packing and hydrophobic interactions. The greater the difference in molecular structure (and hence in intermolecular attractions), the lower the mutual solubility [129]. For this reason, fatty acid-based gelators demonstrate gelling abilities even without the need for substituting functional groups, thereby simplifying the synthesis process.

Simultaneously, the gelation properties of the gelator are influenced by the length of the hydrocarbon chain and the presence of polar functional groups. Indeed, on one hand, polar moieties in the gelator can form bonds with each other through C=O , -OH , amine groups, and aromatic rings, triggering hydrogen bonds and π – π stacking interactions. On the other hand, polar moieties can interact with functional groups potentially present in low-polar solvents, such as fatty acid esters containing a carbonyl group (C=O), further enhancing the interaction between the solvent and the gelling agent [123].

6. Conclusions

Hazardous solvents continue to be extensively used by conservators working on different types of artworks. The removal of degraded coatings often involves the application of volatile toxic substances on the artwork's surface for prolonged periods. During these interventions, conservators are exposed to the inhalation of Volatile Organic Compounds (VOCs), which can lead to long-term health issues, such as carcinogenic, mutagenic, and irritative effects [131]. Low-polar solvents like hydrocarbons are still commonly used in conservation practices, either as pure solvents for low-polar substances such as waxes or in combination with other solvents for more polar materials. Despite efforts to study greener formulations for conservation treatments, the gelation of organic solvents remains a crucial tool for reducing solvent evaporation.

This study explores the application of selected organogelators in the conservation of cultural heritage, focusing on Low-Molecular-Weight Gelators (LMWGs) and their potential to gel low-polar solvents. LMWGs are biocompatible, biodegradable, and cost-effective molecules capable of entrapping both hydrophilic and lipophilic substances based on their chemical structures. They can form organogels with supramolecular structures of fibrils linked through non-covalent forces, immobilizing even low-polar solvents. Some LMWGs could potentially reduce the toxicity of the traditional linear aliphatic hydrocarbons used in conservation, such as Ligroin and deaeromized White Spirit, thereby decreasing the amount of solvent inhaled by conservators. Certain LMWGs can also gel other low-polar linear compounds, such as biodiesel- and oil-based solvents, enhancing the viscosity of greener solvents to minimize their release on the artwork [70].

LMWGs have gained increasing interest in various sectors, including the pharmaceutical, food, and cosmetic industries, due to their chemical–physical features and envi-

ronmental sustainability. The self-assembly can be triggered by different means, causing the formation of weak intermolecular interactions, mainly involving hydrogen bonding, electrostatic interactions, van der Waals interactions, and π - π interactions. Among the examined organogelators, those based on amino acids, sugars, cholesterol, fatty acids, and anthryl and anthraquinone are shown to be potential building blocks to gel low-polar solvents. However, it is important to note that some of these compounds require a synthesis process that may involve the use of non-green substances to trigger the reaction between the compounds.

In addition, the use of organogels on cultural heritage materials requires in-depth analysis of the gel's rheological properties and physico-chemical stability to avoid issues related to the release of gel residues on the artwork or the strong adhesion of the gel to the surface. For this reason, future research should focus on the optimization of LMWGs to facilitate their implementation in the world of conservation.

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