



Confinement of organic solvents in gel matrices for the cleaning of works of art

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Despite the crucial socioeconomic role of cultural heritage, its preservation remains a relatively niche application in science. We show here representative examples illustrating how recent advances in colloid and soft-matter science can support cultural heritage conservation, with emphasis on “green” and sustainable systems designed to confine neat organic solvents or solvents in complex fluids for the cleaning of artworks. Strong connections are highlighted among material design, application strategies, and the underlying physico-chemical processes, to demonstrate how addressing conservation challenges can generate insights transferable to a wide range of scientific fields. Representative artworks are considered, from wall and easel paintings to paper. Several classes of synthetic polymers—such as acrylates, acrylamides, poly(vinyl alcohol), poly(ethyleneimine), siloxanes, and thermoresponsive systems like poly(N-isopropylacrylamide)-based microgels—have demonstrated effectiveness for confining solvents and warrant further investigation. In parallel, a growing body of bio-derived gel materials is being growingly explored, including agar, acrylated gellan, alginate, Funori (combined with halloysites), poly(vinyl butyral), poly-hydroxybutyrate, and, recently, castor oil. The examples and perspectives presented here, which represent the most recent advances in the field and introduce novel developments with the potential to drive scientific progress across multiple sectors, demonstrate that, when guided by a Safe-and-Sustainable-by-Design (SSbD) approach, soft matter and colloids for cultural heritage preservation can transform from an expanding niche field into a source of inspiration with broad, transversal impact, encompassing both theoretical and applied domains.

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Current Opinion in Colloid & Interface Science 2026, 84:102027

This review comes from a themed issue on **Non-aqueous solvents (2026)**

Edited by **Anne-Laure Fameau** and **Romain Bordes**

For complete overview about the section, refer [Non-aqueous solvents \(2026\)](#)

<https://doi.org/10.1016/j.cocis.2026.102027>

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Introduction

Despite the vital socioeconomic importance of cultural heritage, its preservation still represents a niche sector among the possible applications of science. Across the 1950s up to the 1980s, and tracking back to the previous restoration tradition, materials to treat works of art against degradation were borrowed from the polymer, cosmetics, and even food industries [1,2], while most of the scientific research was dedicated to the development of advanced diagnostic techniques [3–5]. Starting from the seminal work of Enzo Ferroni and Piero Baglioni in the 1970s and 1980s, the full potential of colloids and soft matter to produce advanced restoration materials started to be explored, with the first pioneering developments involving the formulation of $\text{Ca}(\text{OH})_2$ nanoparticles and of oil-in-water (o/w) microemulsions to, respectively, reinforce and clean renaissance masterpiece frescoes in Florence (Italy) [6]. Nowadays, examples of colloids and materials science to tailor materials for cultural heritage preservation span from particles [6–8] to coatings [9,10], complex fluids [6,11], gels [12–14], and even sensors [15,16], addressing challenging remedial conservation tasks such as mechanical reinforcement, cleaning and protection of artworks, as well as preventive conservation as in the detection and absorption of pollutants in museums. These findings demonstrate that conservation science can develop original advanced systems, which in turn have the potential to drive and inspire developments in industrial sectors such as pharmaceuticals, detergency, buildings, and agriculture, in a reversed trend with respect to the traditional restoration practice. The link between conservation science and the industry is even more important considering that art conservation has become a hub for developing new materials based on the Safe-and-Sustainable-by-Design (SSbD) approach [17]. This methodology, increasingly adopted and advocated in “green” and sustainable industry [18–20], integrates

safety and sustainability considerations into the early stages of material and product development, minimizing risks to human health and the environment while ensuring high performance and sustainability across the entire life cycle. In this sense, preservation science can well be a driver for economic regrowth. The scope of this contribution is to give representative examples of how the latest research in colloid science and soft matter for the preservation of cultural heritage can contribute to advanced systems, focusing on the confinement of organic solvents to clean works of art. With degradation processes mostly starting at the surface of works of art, cleaning is a recurrent and delicate task [21], where gels and micelles proved the most effective and safe tools to remove soil, grime, aged detrimental coatings, and even vandalism [6]. In this context, confining organic solvents in retentive matrices is the best strategy to clean sensitive target surfaces without affecting original art components, in a noninvasive and time-effective way, surpassing conventional solubilization chemistry. Moreover, gel matrices represent materials of high interest in a wide range of scientific and technological applications [22–25] well beyond art preservation. Starting from the confinement of solvents in micelles and synthetic polymer gels, we will proceed to also cover confinement in gels derived from natural or bio-derived polymers, which represents the frontier in current art cleaning, and besides is pivotal for all the scientific and technological fields where surface interactions drive application performances. Throughout next sections, we will illustrate how the design and application of these materials relate with crucial physicochemical processes, hoping to clearly deliver the concept that addressing these challenges can open for a new insight transferable to different scientific sectors. Different typical artworks will be considered, including iconic wall and canvas paintings, with particular focus on “green” and sustainable restoration systems. Finally, we will provide future perspectives for research in colloids and surface science with high potential in art conservation and beyond.

Confinement of organic solvents in micelles and synthetic polymer gel matrices

Wall paintings, stone, and ceramics

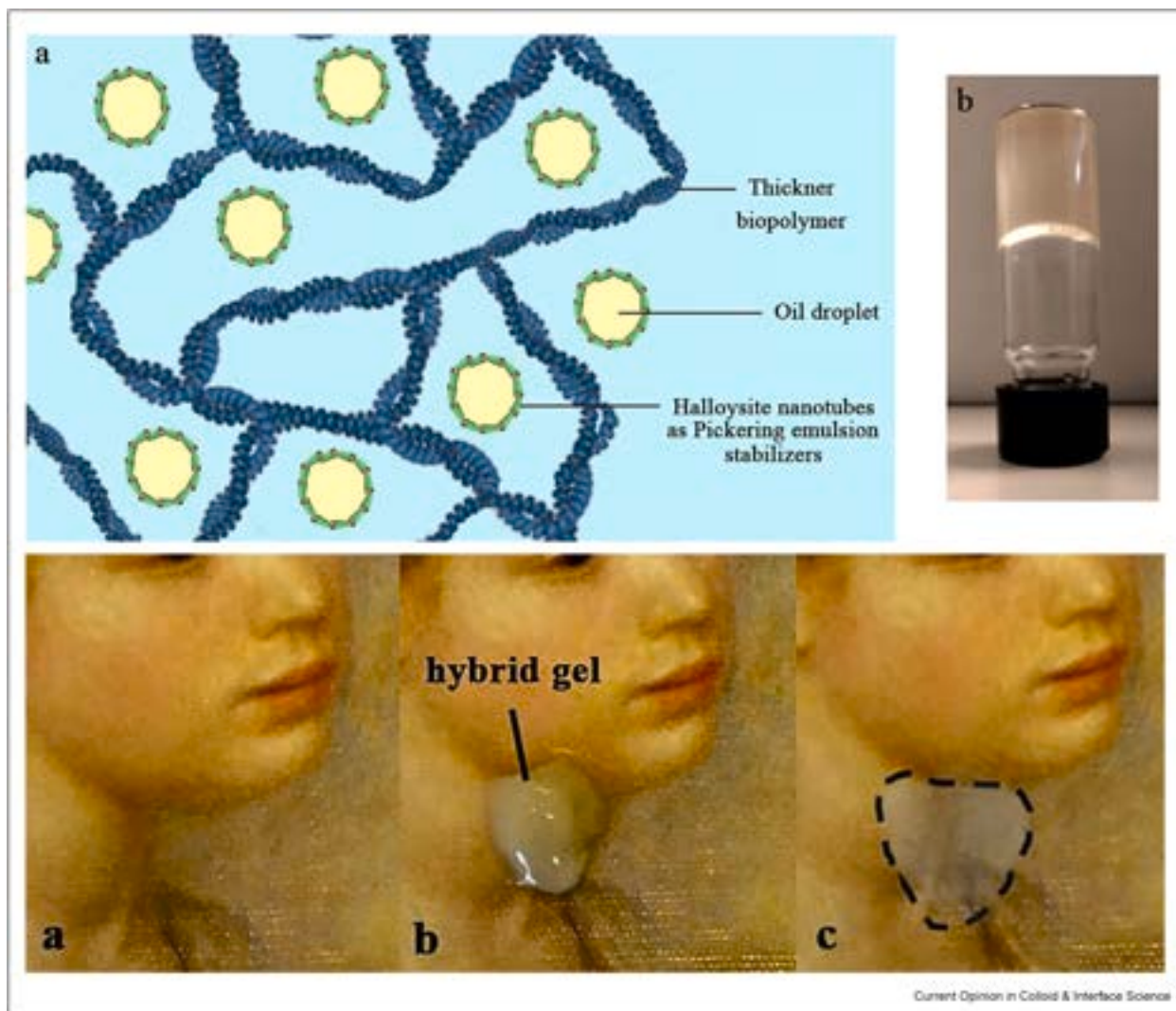
Wall paintings represent an important target when talking about conservation practices involving solvent-based systems as they are ubiquitous in artistic/historical traditions and are often covered by nonpolar coating layers mainly from wrong interventions that alter both the esthetic appearance of the artwork and its physicochemical properties. Essentially, these coatings are hydrophobic and block surface porosities, increasing the crystallization pressure of salts coming out to the surface from the stone bulk, with dramatic consequences and material loss [26]. Indeed, cleaning of jeopardized historical murals is where aqueous o/w nanostructured

systems were firstly tailored and applied to the preservation of cultural heritage starting from the 1980s to the 2000s, with case studies involving the Italian Renaissance (in Florence), and Mesoamerican cultures (in Mexico) [6]. In this case, the organic solvents are partially (if water soluble) or completely (if water insoluble) confined within self-assembled nanosized micelles built by surfactants and cosurfactants, in a water bulk phase [27]. Cosurfactants are added to increase “disorder” in the interfacial film, a concept pioneered by Schulman in the 1940s, yielding highly flexible interfaces with limited persistence length. These o/w liquid systems leverage on different physicochemical process to interact with aged coatings, such as dewetting or controlled swelling/detachment, marking a paradigm shift from traditional polymer thickeners and conventional solubilization chemistry with organic solvents. Several different nanostructured fluids have been confined in cellulose poultices or hydrogels for use on murals.

A systematic comparison between free and confined solvents and microemulsions was carried out by Marazioti et al. [28], who demonstrated that polysaccharide-based gels such as agar and gellan gum effectively control solvent diffusion but require mechanical removal, potentially inducing surface damage. In contrast, microemulsions confined within synthetic hydrogels offered superior performance by enabling controlled softening and dewetting of polymeric layers. Hybrid systems continue to emerge, such as the agar–citrus pectin hydrogel proposed by Spitz-Argolo et al. [29], which showed improved control over the uptake of polar, and protic, solvents, like water and ethanol, compared to pure agar, and the Funori-based gel loaded with halloysite-stabilized Pickering emulsions developed by D’Agostino et al. [30], combining high viscosity, enhanced interfacial activity, and effective removal of hydrophobic resins as depicted in Figure 1. The system may require further improvements since it requires rinsing the treated area with water to remove gel residuals.

Baglioni et al. demonstrated the effectiveness of incorporating alkyl carbonates, a pioneering class of “green solvents” including moderately to highly polar aprotic solvents, into an o/w nanostructured fluid loaded within a hydrogel of poly(2-hydroxyethyl methacrylate)/polyvinyl semi-interpenetrated with pyrrolidone (semi-IPN poly(2-hydroxyethyl methacrylate) [pHEMA]/poly(vinylpyrrolidone) [PVP]) for removing overpaint from street art [31]. This approach enables the use of low-toxicity, environmentally safe fluids and provides a level of cleaning selectivity unattainable with free solvents. The hydrogel was synthesized in water through free radical polymerization, and solvent loading was achieved via solvent exchange. SAXS (Small-angle X-ray scattering) analyses were conducted to elucidate the

Figure 1



Upper panel: (a) scheme of the Pickering emulsion gel based on *Funori* (thickener), haloysite nanotubes (emulsifiers), and isooctane (oil droplets); (b) the system behaving as a gel inside a glass vial. Lower panel: cleaning test using the Pickering emulsion in *Funori* gel. Reprinted adapted with permission from Ref. [30]. Copyright 2024 American Chemical Society.

structure of the microemulsion and the spatial distribution of the solvent within micelles. The application of microemulsion-loaded hydrogels to stone substrates was further explored by Wheththimuni *et al.* [32], who developed a pHEMA/PVP hydrogel loaded with a nanoemulsion based on a nonionic surfactant (EcoSurf™), with polar solvents, both protic and aprotic, such as butanol and butanone, and was employed for the removal of acrylic polymers and copolymers, such as PB72, from two different stone types. The results demonstrated that the gel-based approach outperformed alternative methods, including the use of traditional carriers such as cellulose pulp. Innovative techniques also hold great significance for investigating

solvent diffusion processes in confining matrices. Zhang *et al.* [33] recently proposed a nondestructive bright-field optical imaging strategy to monitor the diffusion of dichloromethane and ethanol, as well as the morphological evolution in individual PMMA (poly(methyl methacrylate)) polymer microspheres. Diffusion dynamics were found to be direction dependent and heterogeneous within single particles, reflecting a nonuniform internal structure. Swelling characteristics could be quantitatively extracted and transient intermediate states captured. The measured kinetics of fully swollen spheres was consistent with predictions from the Li–Tanaka kinetic model for spherical gels, demonstrating the potential of this approach for

resolving complex diffusion mechanisms at the single-entity level. Complementary insights could be obtained by employing different techniques and strategies, adapting approaches such as those proposed by Palombo et al. [34] or Umut et al. [35]

As an alternative to acrylate matrices, Zhou et al. developed a polyvinyl alcohol (PVA)/polyethylenimine physical hydrogel synthesized in dimethyl sulfoxide (DMSO) by dissolving the polymers at 90°C and cooling the mixture in a mold to induce gelation [36]. The resulting gel was loaded with an o/w fluid containing ethyl acetate and propylene carbonate, based on a formulation previously reported by Bonelli et al. [37]. This system was tested for its ability to remove animal glue from a mural painting but also soil from a Cu–Fe metal coin, confirming its versatility across substrates and contaminants.

Another relevant class of confinement matrices for o/w and aqueous cleaning fluids is constituted by polyacrylamide networks. Recent efficient delivery systems are stimuli-responsive microgels based on poly(*N*-isopropylacrylamide) (PNIPAM). Vialletto et al. developed PNIPAM microgels functionalized with oligo-ethylene glycol methyl ether methacrylate surface chains to promote shear deformation through lubrication [38]. Rheological measurements revealed a thermoresponsive inversion of the storage (G') and loss (G'') moduli with increasing temperature. Such tunable rheological behavior resulted to be particularly advantageous for the controlled and complete removal of soiled polymer coatings from textured ceramic surfaces and suggested broader applicability in artistic surface cleaning (see Figure 2).

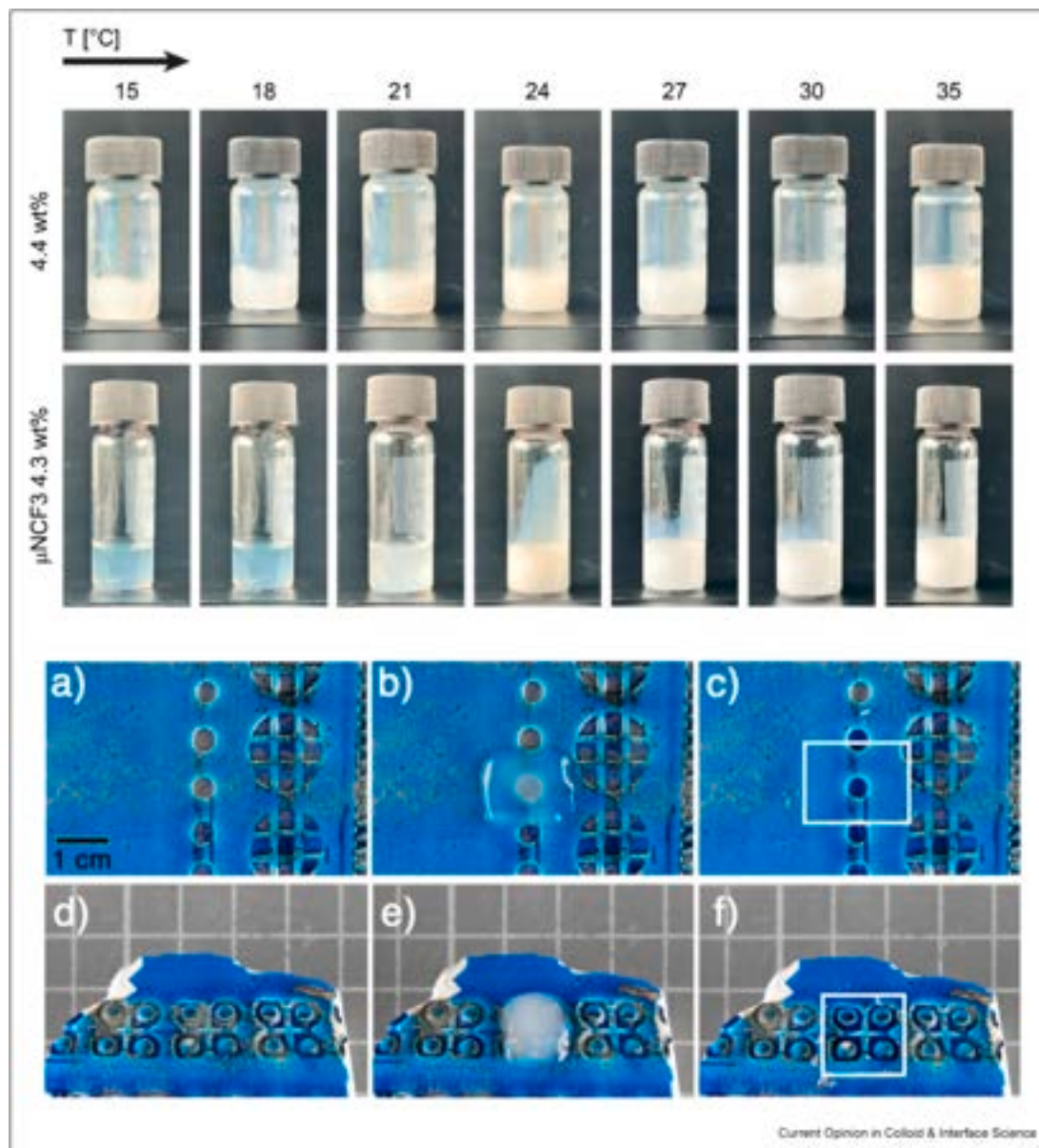
This example finely links art preservation with advanced colloids and surface science, with potential even beyond art preservation. PNIPAM networks are largely relevant materials, whose behavior with confined solvents needs to be further explored. For instance, PNIPAM-based semi-interpenetrating networks were recently examined by Kalkan and Orakdogan to yield systems with enhanced on–off switching and responsive properties [39], a problematic transversal to multiple industrial and technological sectors. By estimating the Flory–Huggins interaction parameter for hybrid solvent systems, the authors demonstrated that semi-IPNs prepared by cold polymerization exhibited anomalous swelling dominated by polymer relaxation, whereas those obtained by warm polymerization followed Fickian diffusion. Adsorption processes were described by pseudo-second-order kinetics, with intraparticle diffusion playing a key mechanistic role. For what concerns acrylamide gels, which had showed potential in confining o/w systems for cleaning artworks [40], complex swelling phenomena were also reported by Toyotama et al. [41], who observed a shrinking–reswelling behavior when they

were initially swollen in water and subsequently immersed in another highly polar, protic solvent, i.e. ethylene glycol. The characteristic times of both shrinking and relaxation were found to scale with the square of the system size, indicating diffusion-controlled dynamics. A coupled diffusion model was proposed to account for this behavior. Gussenov et al. studied polyacrylamide-based gels as oil-recovery systems, examining their swelling behavior in ionized water and mixed water/ethanol or water/acetone solvents [42]. Swelling was mainly diffusion driven and mechanical properties were enhanced by incorporating bentonite clay, allowing control over both Young's modulus and swelling degree.

While the combination of o/w nanostructured fluids with hydrogels currently represents the most advanced standard for the cleaning of works of art, the exploration of new advanced confining matrices for organic solvents is essentially driven by two factors, i.e. the possibility of investigating green solvents with intriguing chemical and physical properties and the need to address a wider range of low-polarity coatings to be removed. One strategy to improve the safety and selectivity of such interventions involves increasing solvent viscosity to limit uncontrolled diffusion. Yu et al. [43] evaluated three commercially available thickening agents—poly(acrylic acid) (Carbopol), agar (Nevek), and a silicate-based material (Laponite RD)—for gelling moderately to highly polar aprotic solvents, i.e. acetone and dimethyl carbonate (DMC), in the removal of a polyvinyl acetate (PVAc) layer from a mural painting in the Payathonzu Temple, collocated in the Bagan archeological site, in Myanmar. Their findings underline the critical relationship among viscosity, solvent mobility, and release dynamics. Lower viscosities enhance solvent diffusion and interaction with the PVAc layer, whereas higher viscosities retard evaporation and enable more controlled release. These effects were assessed through viscosity measurements, weight-loss tests, and PVAc removal assays. Moreover, the intrinsic solubilization ability and volatility of the solvent significantly influence cleaning performance, with DMC outperforming acetone due to its higher affinity for PVAc and slower evaporation. A key point that remains to be addressed concerns the evaluation of posttreatment residues and the risk of physicochemical microalterations of the treated surface. It would be also important to evaluate clearance steps through specific analyses, to rule out possible adverse effects resulting from an insufficiently controlled release.

Moving to solid-like viscoelastic materials for confining solvents, Porpora et al. introduced nonaqueous poly(-dimethylsiloxane) (PDMS) organogel sponges synthesized via a sugar-leaching technique, allowing precise control over porosity and pore size [44].

Figure 2

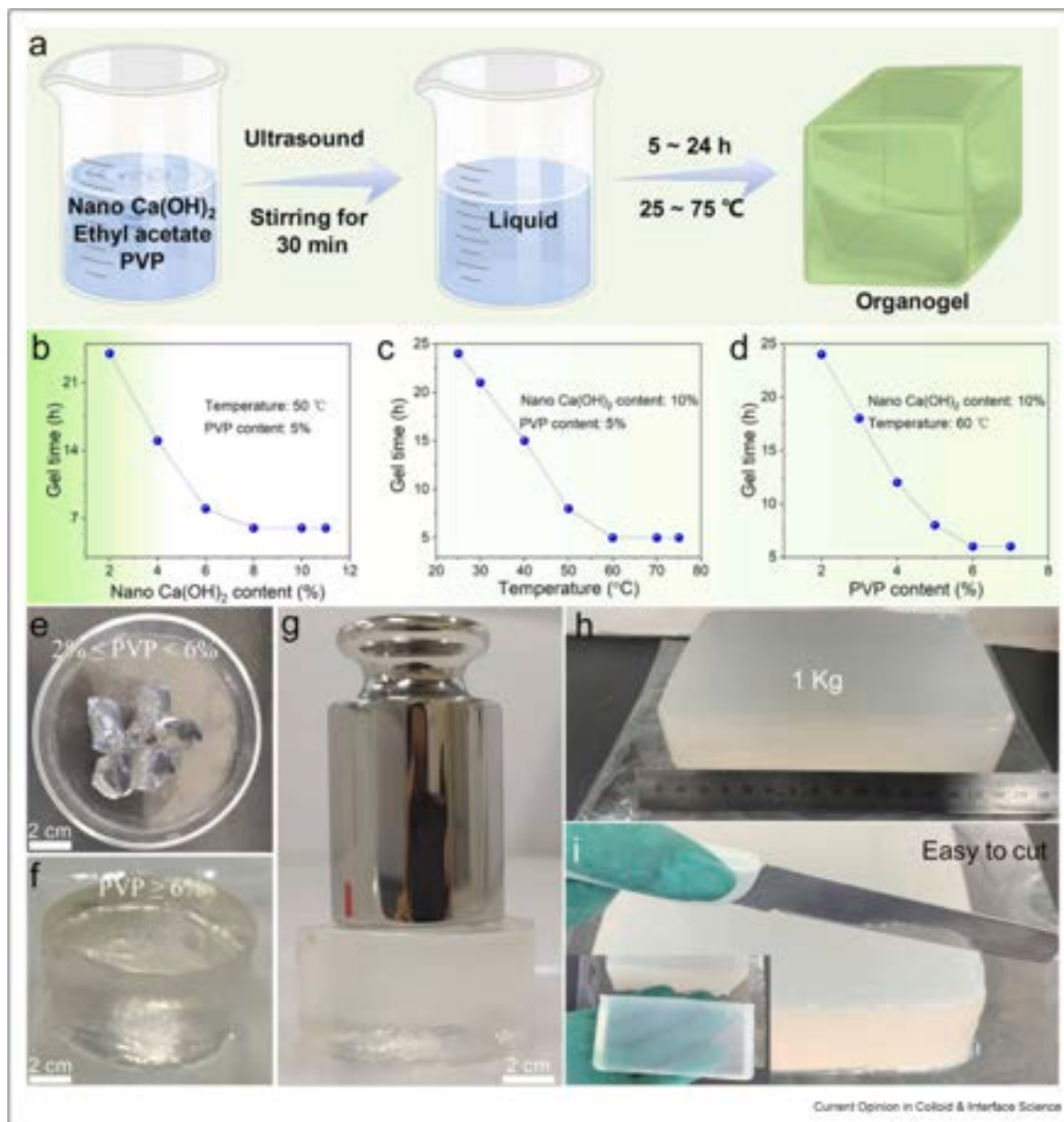


Upper panel: changes in the mixtures in response to temperature are shown in the picture, depending on the composition. Lower panel: application tests of a selected system for the removal of a Plextol B500® layer from a clay sample. Reproduced from Ref. [38]. © Vialeto *et al.*, 2025.

The authors demonstrated that solvent absorption kinetics and capacity are primarily regulated by porosity, with highly porous structures absorbing substantially more solvent. However, no kinetic modeling of solvent uptake was provided. The intrinsic hydrophobicity of PDMS leads to preferential absorption of low-polarity solvents such as cyclohexane. Importantly, the sponges retain their solid-like viscoelasticity after solvent loading and exhibit controlled solvent release, limiting deep penetration into porous substrates such as frescoes or canvas. When applied to mock-up samples, the less

porous PDMS system loaded with acetone achieved selective removal of aged varnishes (ketone resin and polyacrylic copolymer), demonstrating its potential as a residue-free and minimally invasive cleaning tool. A different approach to obtain rigid organogels was proposed by Zhou *et al.*, who developed systems composed of PVP, nanoparticles of $\text{Ca}(\text{OH})_2$, and ethyl acetate [45] as shown in Figure 3. This expands on the potential of PVP to yield polymer networks for art cleaning applications since this compound has been so far largely explored in the formation of semi-interpenetrated

Figure 3



Upper panel: synthesis of an organogel containing PVP. Middle panel: effects of $\text{Ca}(\text{OH})_2$ content, PVP content, and temperature on the gelation of the organogel. Lower panel: pictures of organogels at different PVP contents (e and f), the gel under a 200g weight (g), and the visual aspect of a bigger gel and its shaping (h and i). Reprinted from Reference [45], Construction and Building Materials, 451, Jinmeng Zhu, Jinghan Ding, Yuke Li, Zhang He, Zhenzhen Ma, Wenqiang Dong, Xichen Zhao, Xuanhua Li, Locking organic solvents by crystallization-induced polymer network, 138844, Copyright (2024), with permission from Elsevier. PVP, poly(vinylpyrrolidone).

hydrogel networks to confine aqueous solutions (e.g. chelating agents) or o/w nanostructured cleaning fluids [46,47]. In the work by Zhou et al., phase separation induced during mixing promotes the segregation of PVP from the solvent, thereby facilitating the formation of a polymeric network capable of encapsulating ethyl acetate and ethanol. Concurrently, the reaction between nano- $\text{Ca}(\text{OH})_2$ and ethyl acetate generates calcium

acetate ($\text{Ca}(\text{CH}_3\text{COO})_2$), which forms ionic interactions with the C–O groups in PVP through Ca^{2+} coordination. Due to the poor solubility of $\text{Ca}(\text{CH}_3\text{COO})_2$ in ethyl acetate or ethanol, the salt crystallizes and precipitates within the network. These gels can incorporate a wide range of organic solvents—up to 12 as demonstrated by the authors—and exhibit strong cleaning efficacy on aged polymeric coatings. The work

presents a novel, nonaqueous, environmentally friendly physical gel system suitable for the safe removal of polymeric layers from sensitive cultural heritage surfaces. Jia *et al.* [48] instead propose a shift toward greener systems by developing a gel system that combines the gelling capabilities of PVA with the cleaning efficiency of a green polar aprotic solvent, *i.e.* γ -valerolactone (GVL), which had already showed potential for confinement in the cleaning of historical metals [49]. PVA forms a rigid, fibrous network, while the solvent phase consists of a deep eutectic solvent (DES)—a mixture of choline chloride (ChCl) and ethylene glycol (EG), responsible for physical crosslinking with PVA via hydrogen bonding—and GVL as the primary eco-friendly cleaning agent. Prepared through simple mixing, heating, and cooling, the gel enables a controlled release of GVL, as shown in the ChCl–EG–GVL₂–PVA₂ formulation. This slow, regulated diffusion causes gradual swelling and dissolution of coating layers without deep solvent penetration, and the solid-like structure allows easy removal with minimal residue. The system proved effective for removing aged Paraloid B72™ varnish from both mock-up samples and an actual mural painting. This stands as a particularly interesting application since it combines DES, which are gaining growing interest in multiple technological fields [50,51], with GVL, a natural eco-friendly chemical derived from lignocellulosic and carbohydrates biomass [52]. An interesting future development would be the use of natural DES in similar systems, as they are already promising solvents for the pharmaceutical and other sectors [53]. Therefore, the potential for this class of solvents is vast.

Paintings on canvas and other substrates

Canvas paintings constitute a vast, iconic portion of mankind's artistic/historical heritage, and are notoriously difficult to preserve since soil, aged coatings or vandalism need to be removed from water- and solvent-sensitive painted surfaces, as for instance in most modern/contemporary art [21].

For instance, Casoli *et al.* investigated the interaction between siloxane-based nonpolar aprotic solvents and modern oil paint binders by comparing the effects of cyclomethicone D5 and its potential substitute, dodecamethylpentasiloxane (Si5), applied both in free form and when confined within physical gels [54]. It must be noted that D5 and siloxanes pose exposure concerns, as recently reported [55]. KSG-350Z®, a silicone-based thickener was employed as the thickening agent, and the systems were tested on two modern oil paintings to assess their impact on the paint binder. The study demonstrated that neither siloxane is suitable for the hydrophobic treatment of young oil paints. Moreover, the gelled forms of both D5 and Si5 induced binder interference through leaching phenomena in both artworks, with more pronounced effects

observed on the unvarnished oil painting on paper compared to the varnished painting on canvas. In all cases, the less volatile Si5 resulted in more extensive leaching than D5, highlighting the role of solvent volatility in controlling binder–solvent interactions even when the solvent is gel-confined. This is a highly representative example of the difficulties and concerns associated with solvent-based cleaning in art preservation, and of the potential colloids and surface science hold to remediate.

An important contribution to the understanding of polymer film removal processes was provided by Castel *et al.* [56], who investigated the interaction between ultrathin amorphous polymer films (74 ± 1 nm) and aqueous gelled solutions containing small amounts of good solvent, by means of specular and off-specular neutron reflectometry. Their study elucidated the distribution of heavy water and benzyl alcohol within Laropal A81, a widely used protective varnish in easel painting conservation. Swelling kinetics, interfacial roughness, and film morphology were monitored as a function of temperature and increasing benzyl alcohol concentration in dispersions of Pemulen™ TR-2, a hydrophobically modified acrylic acid copolymer. The addition of small amounts of good solvent for the polymer, induced the formation of water-filled cavities within the varnish layer, which progressively grew over time. While increasing solvent concentration significantly accelerated cavity growth kinetics, raising the temperature above the glass transition temperature had a comparatively limited effect.

In general, even in the case of canvas/easel paintings, confining solvents in nanostructured fluids uploaded in hydrogels, currently represents the most advanced and effective system to work this task. Semi-interpenetrating pHEMA/PVP hydrogels and PVA-based cryogels have emerged as particularly effective. In particular, the group of Baglioni developed fine-tunable PVA-based twin-chain polymeric networks (PVA–TC–PNs) for the removal of soil from an acrylic painting on canvas [12]. The presence of two different PVA polymers in the pre-gel solution triggers polymer–polymer phase separation and demixing that produce blob templates, which after freeze-thawing leave a sponge-like interconnected porosity. The authors demonstrated that both structural and functional properties of these networks can be modulated through functionalization with dicarboxylic acids, such as sebacic acid, leading to a new class of TC–PNs. The decoration of one of the two PVA polymers (the porogen) with the diacids modulates the polymer–polymer phase separation and, consequently, the micron-scale porosity and nanoscale tortuosity in the gel networks. This chemical modification significantly influences fluid uptake and release, where higher tortuosity results in higher residence time of the fluid at the gel-target surface

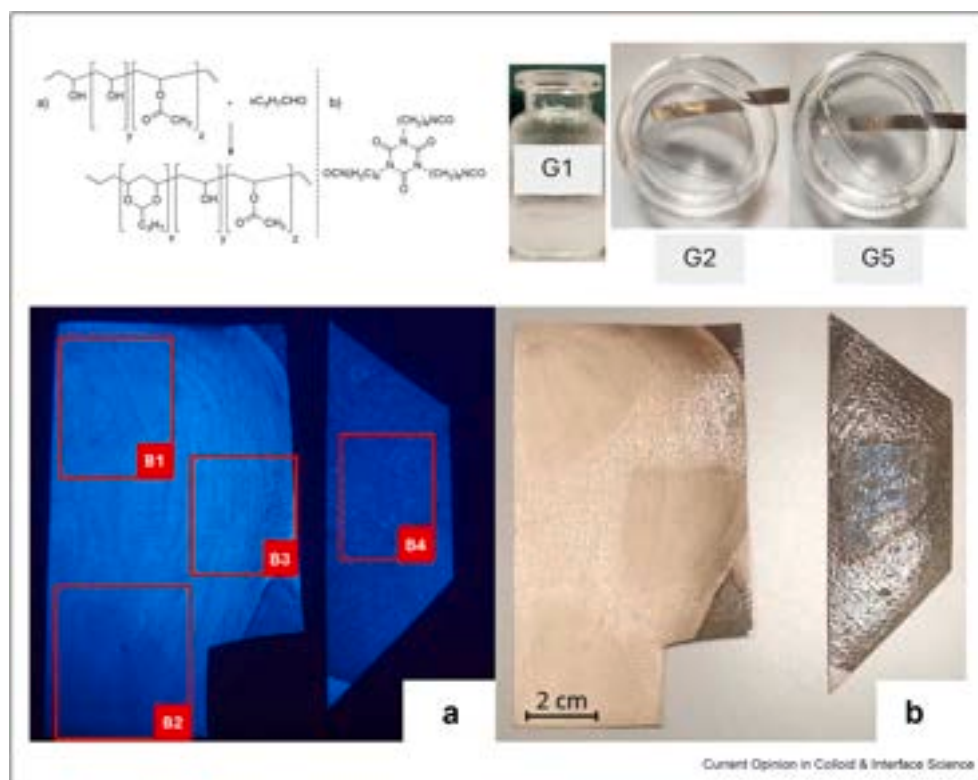
interface, and thus increases cleaning capability. Moreover, the decoration can enhance the affinity for organic solvents with different polarities. The kinetics of porogen and water release were systematically fitted to the Peppas–Korsmeyer equation, and release was found to be predominantly governed by Fickian diffusion, but more investigation is needed to clarify the role of polymer chain swelling in the transport of the fluid through the gel matrix. Nanostructured fluids-loaded TC-PNs have been used so far to restore masterpieces such as works by Pablo Picasso and others [12,57].

Complementary to hydrogels, the design of novel organogel systems is gaining relevance, as alternative tools to address different case studies. For instance, Vespignani et al. [58] developed solid-like gels based on poly(vinyl butyral) (PVB) and isocyanate components, synthesized in DMSO and methyl ethyl ketone (MEK). The synthetic route (see Figure 4) involved initial butyralization of partially hydrolyzed PVA, followed by thermal radical polymerization with Easqua™ M 502, an oligomeric derivative of hexamethylene diisocyanate. These organogels were shown to effectively load environmentally friendly solvents, including MEK, 2-propanol, and DMC. Gel fraction, solvent retention, and release were

quantified using gravimetric methods, and kinetic studies were performed; however, the experimental data were not fitted to specific mathematical diffusion models. The potential for developing organogel matrices from new, sustainable synthetic polymers to confine cleaning solvents is still vast and largely unexplored.

A comprehensive overview of the state-of-the-art approaches to wet art cleaning methodologies was presented by Casini et al., in 2023 [59], including the use of organogels for water-sensitive surfaces. Particular attention was given to polyacrylate-based organogels, for which solvent diffusion mechanisms were analyzed. Solvent uptake was found to follow different combinations of Fickian diffusion and Case II transport, the latter being governed by polymer chain relaxation upon solvent imbibition. The authors also highlighted the increasing incorporation of green solvents and the ongoing transition toward bio-based organogels, such as those derived from polyhydroxybutyrate (PHB), castor oil-based polyurethanes, and triethyl phosphate (TEP). On this topic, Bandelli et al. [60] reported in 2025 an extensive survey of polymers employed in the formulation of organogels for cultural heritage conservation. Polyacrylates, including PMMA, were identified

Figure 4



Upper panel: on the left, the synthesis of polyvinyl butyral (PVB) and the structure of the trimer of hexamethylene diisocyanate; on the right, the visual aspect of the organogels. Lower panel: results of the removal of a dammar resin from canvas mock-ups using the organogels loaded with several organic solvents, under UV light (left) and under visible light (right). Reprinted from Ref. [58] *Progress in Organic Coatings*, 210, Laura Vespignani, Yuewen Pan, Rodorico Giorgi, Antonella Salvini, Polyvinyl butyral organogels for cleaning of artworks, 109730, Copyright (2026), with permission from Elsevier.

as the most used synthetic polymers, whereas castor oil, PHB, and TEP-based systems were recognized as promising candidates for the development of more sustainable gel formulations. Biribicchi *et al.* [61] reported the potential use of low-molecular-weight gelators, such as cholesterol derivatives, fatty acid-derived compounds, anthryl, anthraquinone, amino acid, and saccharide-based organogelators, for low-polarity solvents in artworks cleaning applications. While these systems exhibit promising structuring capabilities, such as self-assembly to form supramolecular structures and high thickening capability at low concentrations, significant challenges remain, particularly in terms of optimizing gel properties and ensuring compatibility with original artistic materials, before their effective implementation in conservation practice.

Confinement of nonaqueous solvents in bio-derived and ‘green’ gel matrices

Canvas paintings

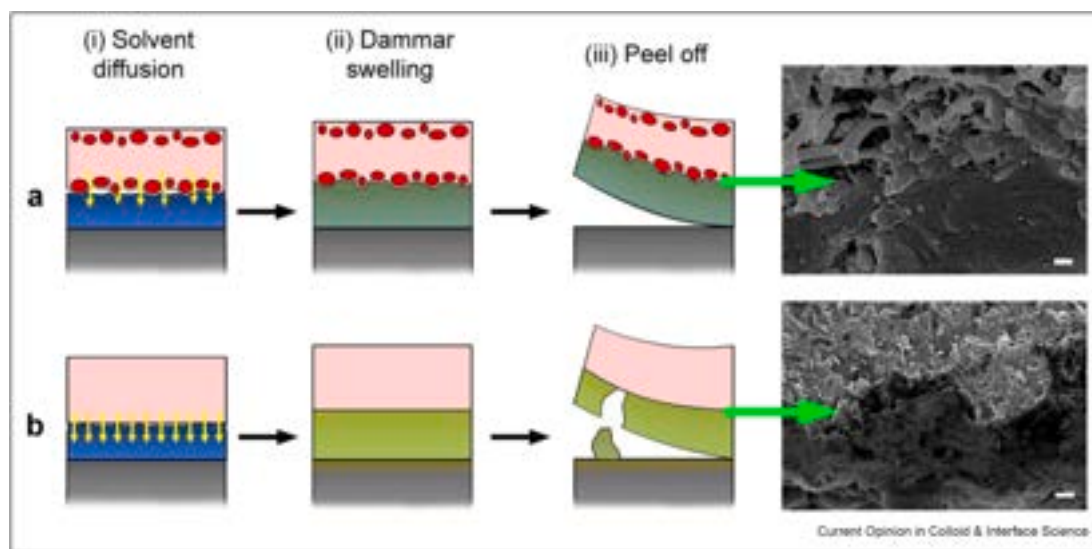
One of the major challenges of recent years in Cultural Heritage conservation is the green transition, which involves the replacement of synthetic, environmentally and toxicologically hazardous materials with bio-derived and more sustainable alternatives. In this context, Passaretti *et al.* [49] provided a comprehensive overview of bio-based cleaning systems developed up to 2021, highlighting the potential of organogels based on biopolymers such as PHB and their combination with sustainable solvents, including GVL, DMC, and a nonpolar solvent, like biodiesel. PHB-based materials have emerged as particularly promising platforms, and the possibility of optimizing their performance to achieve

competitive cleaning systems has attracted significant research interest. In this framework, Yiming *et al.* developed PHB–GVL organogels coupled with electrospun nonwoven layers PVA and polyamide 6,6, demonstrating that these fibrous reinforcements mechanically strengthen the gels and significantly slow down solvent diffusion [62] as shown in Figure 5.

Notably, a higher residual amount of GVL was detected in areas cleaned with PHB–GVL gels alone compared to sandwich composites, suggesting that the presence of fibrous layers reduces the effective solvent release rate by limiting the gel–substrate contact area available for diffusion. Alternatively, Zaratti *et al.* confined fatty acid methyl esters, an emerging class of nonpolar aprotic solvents, within PHB-based gels to remove low-molecular-weight synthetic resins and microcrystalline waxes from canvas mock-ups [63]. Although promising results were obtained, particularly with methyl stearate, the interaction dynamics among solvent, gel matrix, and target resins were not deeply explored.

A different starting point was chosen by Çakmak *et al.* who developed bio-based organogels based on thiol-ene photopolymerized networks derived from isosorbide, pyrogallol, and limonene, swollen with solvents such as DMC, ethyl acetate, and acetone [64]. These gels showed solvent uptake governed by polarity and structural affinity, with DMC exhibiting the highest swelling. Solvent confinement slowed evaporation and limited uncontrolled penetration into paint layers, and cleaning efficiency was attributed to diffusion-controlled solvent release at the gel–varnish interface.

Figure 5



Schematic representation of the mode of action of (a) sandwich-like composites and (b) PHB–GVL organogel for the cleaning of dammar varnish, together with the corresponding SEM (Scanning Electron Microscopy) images. Reproduced from Ref. [62]. Copyright © 2020 American Chemical Society. GVL, γ -valerolactone; PHB, polyhydroxybutyrate.

More recently, Baglioni et al. introduced for the first time castor oil (CO)-based polyurethane networks as solvent-free organogels for cleaning highly water-sensitive artworks [65]. These systems, characterized by SANS (Small-angle neutron scattering) and rheological measurements, are produced and stored as dry gels through a low-energy eco-friendly process with no solvents needed, and then loaded with organic solvents, ranging from nonpolar to polar and including both aprotic and protic (e.g. p-xylene and 1-pentanol) immediately before application. The solvents get confined in the gels' nanosized polymeric mesh. Changing the amount of crosslinking (with biomass-derived isocyanate), the polymeric backbone, and the loaded solvents, it is possible to tune the organization of the gels at the nanoscale, their viscoelasticity, and thus their cleaning capability. These features make the CO organogels highly promising candidates in art cleaning. Subsequent studies focused on performance optimization: Bandelli et al. incorporated oligoesters in the CO gels to fine-tune physicochemical properties and investigated solvent uptake kinetics using the Peppas models [66]. Oligoesters with appropriate $\log P$ values participate in crosslinks and in entanglement regions in the gels, enhancing solvent affinity, and shifting solvent transport behavior from purely Fickian diffusion toward polymer chain relaxation-assisted mechanisms. To support this rational formulation design, Bandelli et al. also proposed a protocol combining SEC (Size Exclusion Chromatography) and DOSY NMR (Diffusion-Ordered Spectroscopy Nuclear Magnetic Resonance) to rapidly determine oligomer hydrophilic–lipophilic balance values [67]. Complementary insight into polyurethane-based gelators was provided by Saharan et al. [68], who studied oil entrapment using polyurethane networks derived from various diisocyanates, interpreting uptake data through Langmuir and Freundlich isotherms and proposing a mechanism based on self-assembly and fiber entanglement forming oil-trapping networks. These gels were tested on substrates such as canvas and wooden paintings, such as a byzantine icon covered with natural varnishes.

Paper

Paper artworks are typically highly challenging to restore and clean, owing to the intrinsic water sensitivity of cellulose fibers to aqueous systems, and of inks/dyes to numerous organic solvents [69,70]. Organic solvents in nanostructured cleaning fluids, confined in hydrogels (pHEMA/PVP or TC PVA networks), have been designed and used successfully by Baglioni et al. to dewet or detach aged adhesives (scotch tapes) from modern paper artworks, while hydrogels simply loaded with water or chelants solutions can remove tidemarks of soil and degradation products from paper [59]. Organogels can be complementary tools in this context, with a focus on bio-derived polymer networks and

“green” organic solvents that show sufficient inertness to inks or dyes.

Richard et al. proposed an innovative strategy to obtain rigid agar-based organogels through a two-step cleaning protocol [71]. The method involves the preliminary application of cyclomethicone D5 to saturate paper pores without solubilizing pressure-sensitive adhesives, followed by the application of an agar-derived organogel obtained through sequential solvent exchange from water to ethanol and finally to methyl ethyl ketone or ethyl acetate. While this approach represents a simple route to prepare a bio-derived polymer organogel, it is not accompanied by a detailed physicochemical characterization of the resulting systems. In addition, D5 and siloxanes pose health exposure concerns as noted in the aforementioned sections. Oliveira et al. developed a gellan gum/PVP gel containing silver nanoparticles, synthesized via gamma irradiation [72]. This system was extensively characterized and shown to uptake both water and organic solvents, such as methanol, ethanol, ethyl acetate, and acetone, while simultaneously releasing Ag^+ ions that provide antimicrobial activity on treated paper and canvas surfaces. An alternative strategy to solvents and microemulsions was proposed by Severini et al. [73], who introduced hydrophobic moieties into methacrylated gellan gum hydrogels to enhance affinity toward organic contaminants such as linseed oil and pressure-sensitive adhesives. In this case, removal is driven not by solubilization but by selective interactions between contaminants and the polymeric network. This concept was further advanced by transitioning from bulk gels to microgels of identical composition [74], increasing specific surface area and improving adaptability to irregular artifact surfaces while enhancing uptake efficiency.

Another highly water-sensitive class of substrates that can therefore require the use of organogels for safe treatment is represented by prints. Ali [75] investigated the suitability of agarose-based gel systems for surface cleaning of albumen prints, systematically evaluating their effects on treated surfaces through a combination of structural and imaging techniques, including SEM (Scanning Electron Microscopy), ATR-FTIR (Attenuated Total Reflectance - Fourier Transform Infrared Spectroscopy), AFM (Atomic Force Microscopy), and colorimetric analysis. The gels were formulated with water and loaded with varying concentrations of different organic solvents, from a polar protic (ethanol) to nonpolar aprotic (toluene). The results demonstrated that agarose-based gels are significantly less aggressive than conventional aqueous treatments typically employed for albumen print cleaning. In particular, free water was shown to induce an increase in crack dimensions, surface roughness, and alterations in the molecular structure of the albumen layer, whereas agarose-based gels loaded with organic

Table 1

Summary of the cleaning systems delivering organic solvents developed during the last five years for treating works of art. The indices next to the solvents indicate their polarity (1: nonpolar, 2: moderately polar, and 3: polar), while the letter specifies whether they are protic (a) or aprotic (b).

Class of polymers	Solvents	Details	Authors
Polysaccharides	Water ^{3,a} , ethanol ^{3,a}	Agar-citrus pectin (AG/CP)-based hydrogel for cleaning an oil painting on brushed aluminum. Absorb water and ethanol in a more controlled manner than AG hydrogels, with better cleaning of painting surfaces.	[29]
Polysaccharides	Isooctane ^{1,b}	Funori-based gel loaded with a halloysite nanotubes Pickering emulsion containing isooctane.	[30]
Polyacrylates, PVP	Alkyl carbonates ^{2-3,b}	pHEMA/PVP-based hydrogel loaded with a nanostructured fluid containing a nonionic CiEj surfactant, an alkyl glycoside, and four different alkyl carbonate green solvents for selective cleaning of contemporary paints versus vandalisms.	[31]
Polyacrylates, PVP	MEK ^{2,b}	pHEMA/PVP-based hydrogel loaded with a nanoemulsion based on EcoSurf™, BuOH, and MEK to remove acrylic polymers, such as PB72 from stone.	[32]
PVA, PEI	Ethyl acetate ^{2,b} , PC ^{3,b} , 1-peOH ^{3,a}	PVA/PEI hydrogels synthesized in DMSO and then loaded with an EAPC-based microemulsion.	[37]
Polyacrylamide, polyacrylates	DEC ^{2,b}	PNIPAM-based microgels with surface oligo (ethylene glycol) methyl ether methacrylate (OEGMA) chains that impart lubricating properties, used as a thermoresponsive delivery system for microemulsions in ceramic surface cleaning.	[39]
Polysiloxanes	Cyclohexane ^{1,b} , DEC ^{2,b} , Ethyl acetate ^{2,b} , 2-propanol ^{3,a} , acetone ^{3,b} , ethanol ^{3,a} , DMSO ^{3,b} , water ^{3,a}	Polydimethylsiloxane (PDMS) organogel sponges used as delivery systems for low-polarity solvents in the cleaning of mural and canvas paintings.	[45]
PVP	Ethyl acetate ^{2,b} , ethanol ^{3,a}	An organogel obtained by blending PVP, nano Ca(OH) ₂ , and ethyl acetate for the confinement of up to 12 different organic solvents to remove polymers from sensitive artworks surfaces.	[46]
PVA	GVL ^{3,b}	New green gels based on choline chloride/ethylene glycol and PVA loaded with GVL as innovative cleaning tool.	[49]
PVA	Water ^{3,a} , DEC ^{2,b} , MEK ^{2,b} , 1-PeOH ^{3,a} , acetone ^{3,b}	PVA-TC-PNs structural and functional properties tuned by functionalizing with sebacic acid. Functionalization affects the porosity and tortuosity of the network, changing its uptake/release of fluids and favoring the uptake of organic solvents with various polarity.	[58]
PVB-co-polyurethane	MEK ^{2,b} , 2-propanol ^{3,a} , DMC ^{2,b}	Organogels based on PVB and isocyanate synthesized in DMSO and MEK. They can load green solvents, such as MEK, 2-propanol and DMC.	[59]
PHB, PVA, polyamide	GVL ^{3,b}	PHB–GVL organogels coupled with electrospun nonwoven layers of poly (vinyl alcohol) and polyamide 6,6 to strengthen the gels and significantly slow down solvent diffusion.	[63]
PHB	FAMES ^{1,b}	FAMES applied through PHB-based gel as green alternatives. Low molecular synthetic resins such as Regalrez 1094 and microcrystalline wax were successfully removed.	[64]
Thiol-ene networks	DMC ^{2,b} , Ethyl acetate ^{2,b} , acetone ^{3,b}	Bio-based organogels based on thiol-ene photopolymerized networks derived from isosorbide, pyrogallol, and limonene, swollen with DMC, ethyl acetate, and acetone. Solvent uptake is governed by polarity and structural affinity, with DMC exhibiting the highest swelling.	[65]
CO-based polyurethane	PeOH ^{3,a} , p-xylene ^{1,b}	Castor oil-based polyurethane networks are innovative sustainable chemical organogels for the cleaning of highly water-sensitive artworks.	[66]
CO-based polyurethane	Ethanol ^{3,a} , acetone ^{3,b} , MEK ^{2,b} , DEC ^{2,b} , p-xylene ^{1,b}	“Green” oligoesters of bio-dicarboxylic acids were used to fine-tune the affinity of castor oil polyurethanes organogels to solvents of different polarity.	[67]
Polysaccharides	Ethanol ^{3,a} , MEK ^{2,b} , ethyl acetate ^{2,b}	Silicone-based solvent cyclomethicone D5 as a treatment aid applied just before the application of a solvent gel, obtained as an agar-based organogels for the delivery of ethyl acetate and MEK.	[72]
Polysaccharides, PVP		A gellan gum/PVP hydrogel containing AgNPs was obtained by polymerization induced by ionizing radiation (γ-rays). These	[73]

(continued on next page)

Table 1 (continued)

Class of polymers	Solvents	Details	Authors
Polysaccharides	Water ^{3,a} , methanol ^{3,a} , ethanol ^{3,a} , ethyl acetate ^{2,b} , acetone ^{3,b}	systems can be loaded with water and various organic solvents, to clean sensitive surfaces while simultaneously providing antimicrobial reservoir.	
Polysaccharides	Water ^{3,a}	Methacrylated gellan gum as an alternative strategy to solvents and microemulsions for the cleaning of paper-based artworks. A transition from bulk gel to microgels of methacrylated gellan gum allowed an increase in specific surface area and an improved adaptability to irregular artifact surfaces.	[74]
Polysaccharides	Water ^{3,a} , ethanol ^{3,a} , toluene ^{1,b}	Agarose-based gel systems to clean albumen surfaces.	[75]
Polysaccharides	Ethyl alcohol ^{3,a} , toluene ^{1,b} , acetone ^{3,b}	Agar and Klucel G loaded with organic solvents (ethyl alcohol, toluene, or a mixture of toluene and acetone) to remove PSTs.	[76]
			[77]

DMSO, dimethyl sulfoxide; EAPC, ethyl acetate and propylene carbonate; FAME, fatty acid ethyl esters; GVL, γ -valerolactone; MEK, methyl ethyl ketone; PC, propylene carbonate; 1-PeOH, 1-pentanol; DEC, diethyl carbonate; PEI, polyethylenimine; PHB, polyhydroxybutyrate; PNs, polymeric networks; PVA, polyvinyl alcohol; pHEMA, poly(2-hydroxyethyl methacrylate); PVP, poly(vinylpyrrolidone); TC, twin-chain; PST, Pressure-Sensitive Tape.

solvents provided a markedly safer and more controlled cleaning approach. In a complementary study, Waheeb et al. [76] explored the removal of pressure-sensitive tapes from prints using agar- and Klucel G-based gels loaded with organic solvents, including ethanol, toluene, and a toluene–acetone mixture. In both cases, solvents were incorporated into the sol–gel systems at controlled concentrations prior to casting, and gels loaded with ethanol exhibited the most effective and selective cleaning performance. Since these systems generally exhibit less-controlled solvent release and could potentially leave residues, their use on solvent-sensitive artworks (e.g. prints with modern inks) should be evaluated carefully.

However, despite the extensive literature on polysaccharide-based gels for conservation, studies explicitly addressing diffusion mechanisms remain scarce. In this respect, the work of Soleimanpour et al. [77] represents a valuable reference, investigating the diffusion of a poorly water-soluble flavonoid, genistein, within mesoporous starch. Adsorption data were best described by the Langmuir isotherm, indicating diffusion-driven monolayer adsorption. Similarly, Lofty and Vivian [78] studied lignin–chitosan nanocomposite hydrogels for fertilizer release, with kinetic analysis revealing combined Fickian and quasi-Fickian diffusion. A polysaccharide of interest for conservation studies that needs further exploration is alginate. Smaniotto et al. offered insight into the use of alginate fluid gels (AFG) as encapsulation media for both hydrophilic actives, such as nicotinamide and hydrophobic ones (vanillin and tryptophan) [79]. The rate and extent of hydrophobic active loading in the AFG particles were governed by the concentration of the former and gel particle size. Release studies under sink conditions revealed that hydrophobic active content within the AFG particles is not expelled and remains fully entrapped regardless of

fluid gel particle size and storage period. Moreover, a hypothesis suggesting that the loading of hydrophobic actives within the AFG particles is due to changes to the solvent quality of water (i.e. the capability of water to solubilize molecules) is presented. Finally, Kumar et al. demonstrated anomalous transport behavior in Ca^{2+} -crosslinked alginate– TiO_2 hydrogels arising from coupled diffusion and polymer relaxation [80].

Overall, the potential for further developments of bio-derived hydrogels and organogels is vast, with the need of matching formulation efforts with advanced physicochemical characterization not just of the gels' structural features but also of their effects on, and relationships with, the transport of solvents and dissolved/detached matter in the gels' networks. In particular, finely decoupling diffusion from chain swelling or adsorption should be addressed to select the best-performing gel materials and solvents.

Table 1 provides an overview of relevant studies published over the last five years focusing on the development of cleaning systems for delivering organic solvents in the cleaning of works of art. As can be observed, polysaccharide gelators show good affinity for polar protic solvents; however, in some cases, these systems are also able to confine moderately to low-polarity aprotic solvents, such as toluene. Synthetic polymers such as polyacrylates and PVP are mostly used to thicken or confine moderately polar aprotic organic solvents, such as alkyl carbonates or MEK, apart from water, whereas PVA and polysiloxanes exhibit greater versatility, being able to gel solvents of different nature. In general, confinement is favored when there is good complementarity between the solvent polarity and the functional groups of the polymer; however, some systems can adapt to very different conditions, highlighting the complexity of gelation phenomena.

Conclusions and future perspectives

Research in colloids, soft matter, and materials for cultural heritage preservation has grown over the last decades to become a vibrant scientific field with a potential for innovation on a much wider scale than its current applications. Key research lines involve the design of systems to confine organic solvents to remove dirt and other unwanted layers from the surface of sensitive works of art. “Green” chemistry and sustainable formulation processes are necessary to promote socioeconomic growth through these developments. In particular, while hydrogels based on semi-IPN or “twin-chain” networks have already shown great efficacy, there is much to explore regarding synthetic or bio-based organogels able to confine “green” organic solvents. Acrylates, acrylamide, PVA, PEI, siloxanes, and PNIPAM (e.g. for microgels) stand as possible synthetic polymer classes that already exhibited efficacy and can be further explored. Among bio-derived gel materials, we have mentioned agar, acrylated gellan, alginate, Funori (with halloysites), PVB, PHB, and more recently castor oil, which can be crosslinked with biomass-derived isocyanates to form bio-based polyurethane networks capable of confining solvents for cleaning art. The possibilities are vast and this contribution’s hope was to show how inspirational soft matter for art preservation can be to devise new gelled systems.

Future perspectives on these systems must also focus on current limitations concerning possible drawbacks in their application.

A primary concern is the risk of residual contamination of artworks by the cleaning systems themselves. Components such as polymers, surfactants, or additives, if released from the gel matrices, may adsorb onto, or partially penetrate inside, the substrate, making their complete removal difficult. This issue is particularly relevant for solvent confinement using thickeners and certain physical gels. In principle, despite the apparent macroscopic effectiveness of the cleaning process, molecular-level alterations might still occur either from the presence of residuals or from alterations of the artistic surfaces by components in the cleaning systems. Such changes can affect the physicochemical state of the surface and underlying layers and potentially influence subsequent conservation treatments or aging processes of the treated works of art.

For these reasons, the design of solvent-confining systems in art preservation is still an open field. Current research is moving toward the development of more robust gel structures, capable of minimizing their release onto treated surfaces during application without sacrificing surface adhesion, mechanical compliance, and the capacity to exchange cleaning fluids at the gel-target surface.

Materials design for cultural heritage preservation, guided by SSbD approach, has the possibility to link with several technological sectors beyond art conservation. For instance, links exist between new gelled materials of interest in pharmaceuticals, cosmetics and healthcare, and art cleaning. In addition to the “twin-chain” PVA gels, composite polyelectrolyte gels are also a possible class of interest for future developments. Kubra *et al.* designed semi-IPN nanocomposite cryogels based on anionically modified polyacrylamide and a fibrous nanofiller sepiolite [81]. Detailed swelling studies showed that, at alkaline pH, water sorption was primarily diffusion-controlled as indicated by diffusional exponents of $n < 0.5$. Similarly, Cui *et al.* employed polyampholyte gels based on p-styrenesulfonate and N,N-dimethylaminoethyl acrylate to investigate swelling and shrinking kinetics, revealing that swelling is governed by the permanently crosslinked network, whereas shrinking is controlled by structural frustration arising from large differences in the heat and solvent diffusions [82].

Similarly, decorated silica aerogels represent an intriguing class of matrices. Cok *et al.* developed vinyl- and methyl-decorated silica aerogel-like hybrids and correlated their physicochemical and microstructural properties with oil adsorption performance [83]. Increasing hydrophobicity enhanced adsorption capacity, while higher vinyl content led to reduced total pore volume and densification of the silica network, shifting the structure from microporous/mesoporous to predominantly macroporous. Functionalized mesoporous silica was also investigated by Yamada *et al.* [84], who examined solvent-dependent mass transfer of Coumarin-102 in octadecylsilyl-modified silica gels. In acetonitrile/water and DMF (dimethylformamide)/water mixtures, the formation of organic-rich interfacial layers suppressed pore diffusion, resulting in non-Fickian, biexponential uptake kinetics. Conversely, in butanol/water systems, pore accessibility was maintained and transport approached Fickian behavior. These results demonstrate that solvent-induced interfacial structuring critically governs mass transport in hydrophobic mesoporous networks. Despite the versatility and widespread use of polyethylene glycol (PEG), PEG-based gels have not been recently reported as cleaning tools in cultural heritage conservation. Nevertheless, an interesting study by Zahra *et al.* described PEG-based hydrogels exhibiting pH-dependent swelling behavior, with release kinetics governed by solvent diffusion mechanisms [85]. Finally, for what concern bio-derived polymers, tartaric acid-derived cellulose-based hydrogels appear particularly promising. Tan *et al.* reported cellulose hydrogels modified with tartaric acid exhibiting non-Fickian initial swelling followed by Schott second-order kinetics and polymer chain relaxation [86].

These perspectives highlight the vast potential of art conservation research to move from an expanding niche into an inspirational sector with transversal impact in multiple colloid and surface science theories and practical applications. In this context, the development of novel gel-based materials for cultural heritage preservation and beyond emerges as a flourishing research area, driven by the interplay among chemistry, physics, materials science, and conservation practice. Key material features—including stiffness, microstructure and nanostructure, and diffusion properties—play a central role in tailoring performance for specific applications. However, the field still lacks a comprehensive development protocol that effectively balances greener, sustainable synthesis with the systematic adoption of advanced analytical methodologies. Establishing such an approach would not only represent a significant advance in cultural heritage conservation but would also result in major breakthroughs in surface-related technologies.

Credit authorship contribution statement

Leonardo Bellandi: Conceptualization, Writing – review & editing, Writing – original draft; Damiano Bandelli: Conceptualization, Writing – review & editing, Writing – original draft, Supervision; Giovanna Poggi: Writing – review & editing, Writing – original draft, Validation; David Chelazzi: Conceptualization, Writing – review & editing, Writing – original draft, Validation, Supervision, Funding acquisition; Piero Baglioni: Conceptualization, Writing – review & editing, Validation, Supervision, Funding acquisition.

Declaration of competing interest

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

Acknowledgements

CSGI and the European Commission (GREENART project, Horizon Europe research and innovation program under grant agreement no. 101060941) are gratefully acknowledged for financial support. Views and opinions expressed are however those of the author(s) only and do not necessarily reflect those of the European Union or the European Research Executive Agency (REA). Neither the European Union nor the granting authority can be held responsible for them. DC and DB acknowledge the Project PE 0000020 CHANGES – CUP [SPOKE 7, WP5], NRP Mission 4 Component 2 Investment 1.3, funded by the European Union–NextGenerationEU for financial support. The publication was made by a researcher (DB) with a research contract funded by the European Union–PNRR NextGenerationEU in accordance with Article 24, paragraph 3a of Law No. 240 of December 30, 2010,

as amended and Ministerial Decree No. 1365 of November 8, 2022.

Data availability

Data will be made available on request.

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This work demonstrated the potential of polydimethylsiloxane PDMS sponges to confine organic solvents in the selective cleaning of works of art. The role of pore size in the PDMS elastomeric network is studied as related to solvent uptake and release, opening for a new potential class of solvent-confining systems.

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corrosion removal. *ACS Appl Mater Interfaces* 2022, **14**:7471–7485, <https://doi.org/10.1021/acsami.1c22837>.

Novel networks semi-interpenetrated with poly(acrylic acid) (PAA) or poly(vinylpyrrolidone) (PVP) are formulated to capture copper ions and remove bronze corrosion patinas. The gels are characterized through chord length analysis, small-angle X-ray scattering and FTIR Imaging. Chlorides from patinas dissolve and migrate inside the gels, where Cu(II) ions form ternary complexes with chelants and carboxylates in PAA or carbonyls in PVP. The PHEMA/PAA gels have pH-responsive behavior appealing also to drug delivery, wastewater treatment, etc.

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