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Hot Paper

Hydrophobic Eutectogels for the Removal of Contaminants of Emerging Concern from Water

Elena Gabirondo,[a, b, c] Joao M. M. Araújo,[b] Ana B. Pereiro,[b] and Liliana C. Tomé*[a, b]

Five hydrophobic eutectic solvents based on menthol and thymol were prepared and incorporated into a poly(ethylene glycol) diacrylate (PEGDA) network to form eutectogel membranes. The two most promising eutectogels were further optimized by adding ethyl hexylacrylate (HA) to the eutectogel formulation to improve the compatibility between the eutectic solvent and polymer network and to enhance the water resistance of the resulting membranes. Thermal analysis confirmed the successful formation and integration of eutectic solvents within the polymer network. Rheological studies demonstrated the rubber-like behavior of the prepared hydrophobic eutectogels, with menthol-based variants exhibiting superior mechanical properties. Finally, sorption experiments were conducted using the optimized octanoic acid:menthol PEGDA-HA eutectogel to evaluate its efficiency in removing various contaminants of emerging concern (CECs), including diclofenac, iopromide, cefazolin, bisphenol A, and dichlorophenol. The results revealed high sorption capacities for bisphenol A (3213 mg·kg^{-1}) and dichlorophenol (2981 mg·kg^{-1}), followed by diclofenac (1490 mg·kg^{-1}), whereas minimal sorption capacities were observed for iopromide and cefazolin. Overall, this study demonstrates the potential of hydrophobic eutectogels as efficient and tunable materials for water purification, paving the way for their application in the environmental remediation of different emerging pollutants related to global change and human activities.

Introduction

Among the United Nations Sustainable Development Goals (SDGs) established to tackle the world's most pressing challenges,[1] Goal 6 stands out for its focus on ensuring access to clean water and sanitation.[2] While significant efforts have been made in improving water quality, the presence of emerging contaminants, such as pharmaceuticals, contrast agents, hormone-disrupting chemicals, pesticides, and cosmetic products, remains a growing concern in industrialized countries.[3–8] For instance, pharmaceuticals enter water systems primarily through human excretion and unregulated discharge.[9] This poses a clear threat to both the environment and public health.[10]

Various water treatment technologies, such as filtration using activated carbons and advanced oxidation processes, have been explored to remove contaminants from drinking water.[11,12] Activated carbon filtration relies on porous carbon materials to adsorb organic matter, pesticides, and other contaminants,[13–16] whereas oxidation processes use powerful oxidizing agents to decompose complex organic pollutants into simpler, less harmful compounds.[17–19] Despite their effectiveness, these methods often face limitations such as high operational costs or incomplete removal of certain compounds.[20,21] Therefore, as water quality standards become more stringent and new contaminants of concern emerge, there is an urgent need for innovative materials to develop advanced technologies or enhance existing ones.

Eutectic solvents (ES) have emerged as promising alternatives to their predecessors' ionic liquids in the development of soft materials, opening the door to the creation of eutectogels as sustainable alternatives to iongels for diverse applications.[22–24] In this context, hydrophobic eutectogels represent a novel class of soft materials that combine the properties of hydrophobic eutectic solvents with the structural integrity of solid-like materials. These innovative materials can be prepared by embedding hydrophobic eutectic solvents into polymeric networks, resulting in a distinctive combination of properties that are advantageous for water-based applications. Despite being a recent approach, hydrophobic eutectogels have been attracting increasing attention. Studies have already demonstrated their potential for applications such as underwater sensing,[25–27] and drug delivery systems.[28]

The hydrophobic nature of these eutectogels allows them to repel water while selectively interacting with certain organic compounds, making them promising candidates for environmental remediation, such as the removal of contaminants from water.[29] Their gel structure provides mechanical stability, facilitating easy handling and recovery after use. Furthermore,

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[a] CEMMPRE, ARISE, Department of Chemical Engineering, University of Coimbra, Coimbra, Portugal
[b] LAQV, REQUIMTE, Department of Chemistry, NOVA School of Science and Technology, FCT NOVA, Universidade NOVA de Lisboa, Caparica, Portugal
[c] POLYMAT and Department of Polymers and Advanced Materials: Physics, Chemistry and Technology, Faculty of Chemistry, University of the Basque Country UPV/EHU, Donostia-San Sebastian, Spain

Correspondence: Prof. Liliana C. Tomé, CEMMPRE, ARISE, Department of Chemical Engineering, University of Coimbra, Rua Sílvio Lima, Coimbra 3030-790, Portugal.
Email: liliana@eq.uc.pt

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the variety of starting materials and possible combinations to prepare the eutectic solvent component offers a versatile tool for precisely controlling the physical and chemical properties of eutectogels, enabling the development of tailor-made materials for specific target pollutants in water treatment processes.

In this study, hydrophobic eutectogels were developed to remove contaminants from water, particularly pharmaceuticals. Five eutectic solvents based on menthol and thymol were initially prepared and incorporated into a polymeric acrylic network to obtain membranes. The chemical and physical properties of the resulting membranes were characterized to evaluate their suitability for underwater applications. The two most promising membranes were further optimized to improve their water resistance. Finally, the performance of the selected eutectogel membrane was assessed based on its ability to remove various contaminants of emerging concern (CECs), including diclofenac, iopromide, cefazolin, bisphenol A, and dichlorophenol.

Materials and Methods

Materials

The compounds used for the preparation of eutectic solvents, including tetrabutylphosphonium chloride ($P_{4444}Cl$), n-benzyltributylammonium chloride ($N_{bz444}Cl$), choline chloride (ChCl), octanoic acid (C8), thymol (Thy) and menthol (Men), were purchased from Sigma Aldrich and used as received. The acrylates poly(ethylene glycol) diacrylate (PEGDA, $Mn = 575\ g \cdot mol^{-1}$), 2-hydroxyethyl methacrylate (HEMA), ethylene glycol dimethacrylate (EGDMA), dipropylene glycol diacrylate (DPGDA), ethyl hexylacrylate (HA), and the photoinitiator, 2-hydroxy-2-methylpropiophenone (Darocur 1173, 97%), were also supplied by Sigma Aldrich. Contaminants diclofenac sodium salt, dichlorphenol, and iopromide were purchased from Sigma Aldrich, while cefazolin sodium salt and bisphenol A were supplied by TCI.

Preparation of Eutectogels

The eutectic solvents were prepared by combining hydrogen bond donors (HBD) and hydrogen bond acceptors (HBA) at specific molar ratios. The mixtures were heated to 70 °C for 1 h, then allowed to cool to room temperature. Various HBA:HBD molar ratios were tested, and only combinations that remained liquid at ambient temperature were selected for further study. For $P_{4444}Cl$-Thy, $Nbz_{444}Cl$-Thy, and ChCl-Thy, the optimal HBA: HBD ratio for forming stable eutectic mixtures was 1:4. In contrast, the optimal ratio was 2:1 for both C8-Thy and C8-Men systems.

Eutectogels were obtained by mixing eutectic solvents (60 wt%) and acrylates (40 wt%) with a photoinitiator (5 wt% relative to the acrylates). For formulations using HEMA as the gelator, 5 wt% of EGDMA was added to 35 wt% of HEMA. The resulting mixtures were poured into silicone molds and exposed to UV irradiation at a wavelength of 365 nm for 10 min. After curing, the eutectogels were removed from the molds and stored at room temperature for subsequent use.

^{1}H NMR Spectroscopy

^{1}H Nuclear Magnetic Resonance (^{1}H NMR) spectra were recorded on a Bruker Avance DPX 300 at 300.16 MHz, using deuterated dimethylsulfoxide (DMSO-d_6) as the solvent at room temperature.

FTIR Spectroscopy

Attenuated total reflectance Fourier-transform infrared spectroscopy (ATR-FTIR) was performed using a Perkin Elmer Spectrum Two Spectrometer. All the spectra were collected using 12 scans from 400 to 4000cm^{-1}.

Contact Angles

The contact angles (θ) were measured using water, formamide, ethylene glycol, and dioodomethane with the KSV Cam 100 (KSV Instruments LTD, Helsinki, Finland) software at room temperature. Each contact angle value was the average of three measurements acquired immediately after drop deposition. These values were used to calculate the polar and dispersive components of the surface energy of the eutectogels using the Owens–Wendt approach.[33]

Differential Scanning Calorimetry

Differential Scanning Calorimetry (DSC) was performed using a PerkinElmer 8500 DSC instrument. Non-recyclable aluminium hermetic pans were used, and the analyses were performed under a nitrogen atmosphere. The heating and cooling cycles were conducted at 10 °C·min^{-1}. For the eutectic solvents, the samples were heated to 100 °C, whereas the eutectogels were analyzed by heating to 150 °C. Second-run heating cycles were recorded to investigate the phase transition behavior of all samples, ensuring the removal of any thermal history effects.

Thermogravimetric Analysis

Thermogravimetric analysis (TGA) was performed using a PerkinElmer Thermogravimetric Analyzer. The samples were heated to 600 °C at a rate of 10 °C·min^{-1} under a nitrogen atmosphere to ensure thermal decomposition analysis in an inert environment.

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Rheological Studies

The rheological properties of the eutectogels were assessed using a Kinexus Prime Pro+ rheometer (Netzsch) equipped with a plate-plate geometry. The temperature was maintained at 25 °C throughout the experiments. Cylindrical eutectogel samples with a diameter of 10 mm and a thickness of 1 mm were used. Strain sweeps were conducted over a range of 0.01 % to 500 % at a fixed frequency of 1 Hz, while frequency sweeps were carried out from 50 Hz to 0.01 Hz at a constant strain of 0.5 %.

Scanning Electron Microscopy

Scanning Electron Microscopy images of the surface and cross section of the prepared eutectogel were taken with an Analytical TableTop Microscope/Benchtop SEM TM3030 (Hitachi, Japan), equipped with a field emission gun operated at 15 kV. All samples were coated with a thin Au layer to induce charge under the electron beam.

Sorption Experiments

Prior to the sorption experiments, the eutectogels were immersed in water for 24-hour to remove any non-incorporated eutectic solvent and residual monomer. The adsorption kinetics were performed at 100 rpm and 25 °C using dried eutectogel disc samples weighing approximately 65 mg. Each disc sample was submerged in 5 mL of an aqueous solution containing 40 mg·mL^{-1} of the target contaminant. After the adsorption experiments, 3 mL of each sample was analyzed using a VWR P6 UV/Vis Spectrophotometer. Calibration curves were prepared for each contaminant, and absorbance measurements were recorded at the following wavelengths: 276 nm for diclofenac, 242 nm for iopromide, 271 nm for cefazolin, 284 nm for dichlorophenol, and 276 nm for bisphenol A. All experiments were performed in triplicate, and the average values were reported.

Results and Discussion

A set of five hydrophobic eutectic solvents was prepared using thymol and menthol as hydrogen bond donors, combined with various hydrogen bond acceptors (Figure 1a), including ChCl (choline chloride), $P_{4444}Cl$ (tetrabutylphosphonium chloride), $N_{bz444}Cl$ (n-benzyltributylammonium chloride), and C8 (octanoic acid). Thymol and menthol were selected because of their well-known hydrophobic properties. Except for ChCl, the hydrogen bond acceptors were primarily chosen for their long hydrocarbon chains, which are anticipated to further enhance the hydrophobicity of eutectic solvents.

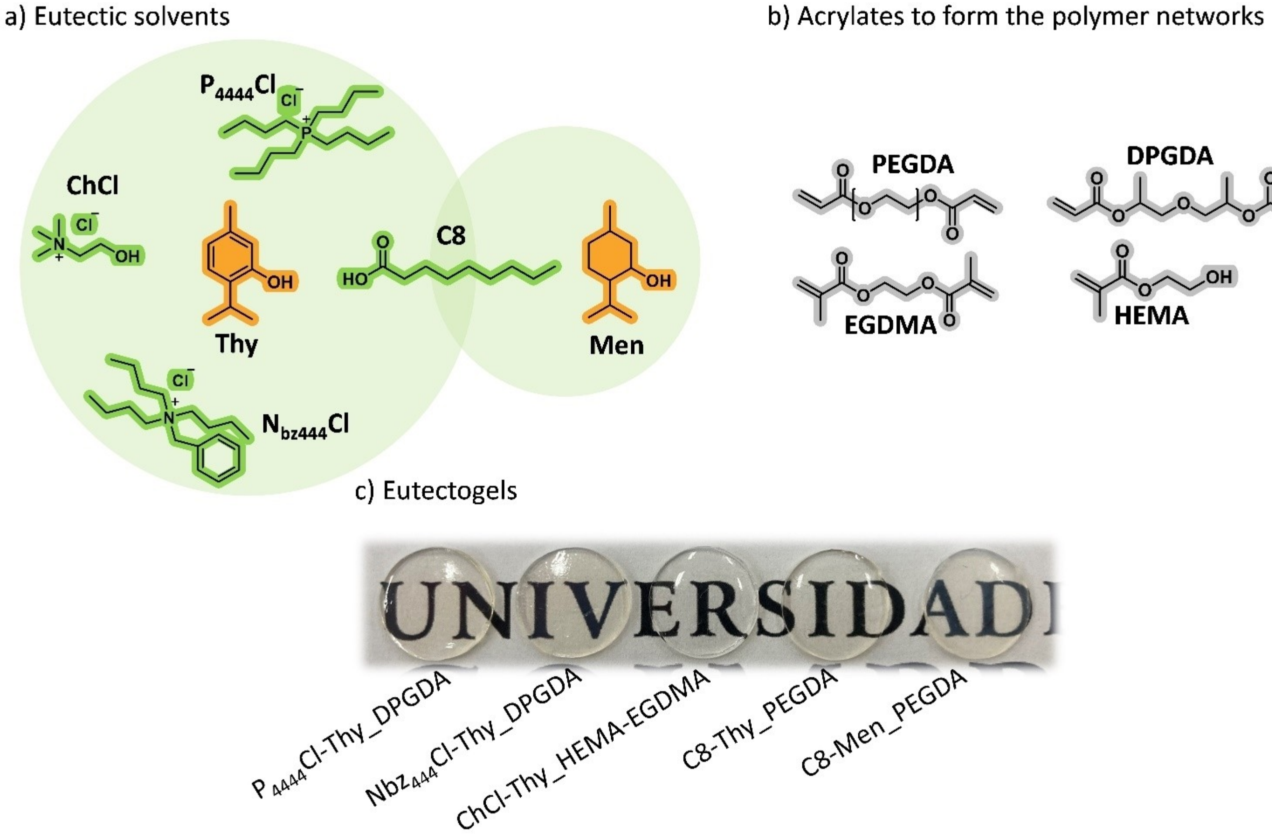

Figure 1. Structures of a) the eutectic solvent components, b) acrylates to form the polymer networks, and c) photo of the prepared eutectogels.

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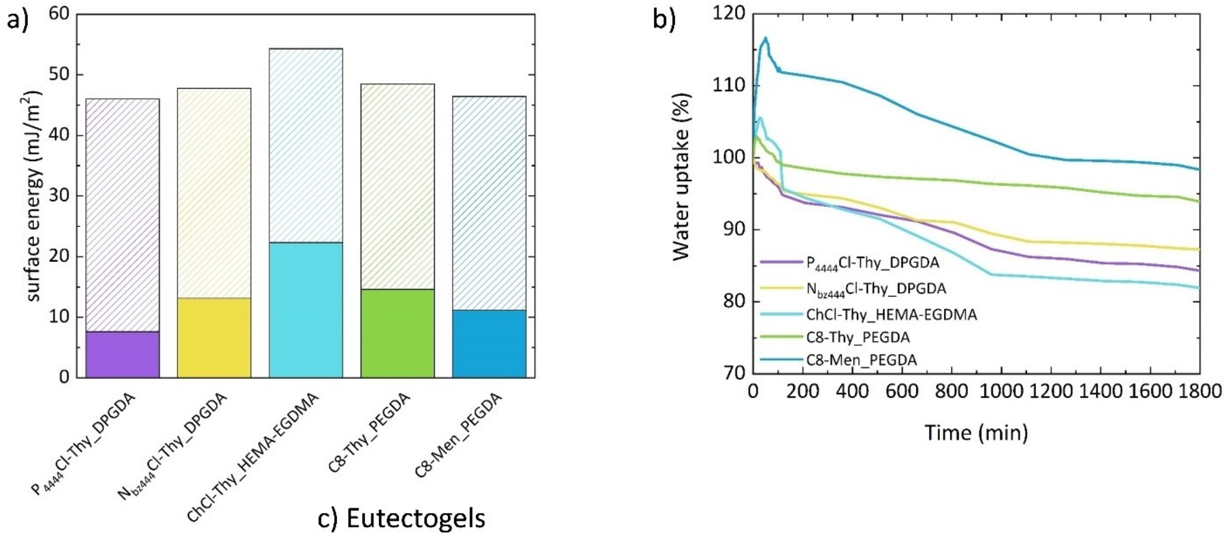
The formation of the eutectic solvents was confirmed by DSC (Figure S1), ^{1}H NMR (Figures S2–S6) and FTIR (Figures S7–S11) analyses. From the DSC analysis it can be observed that there was a depression of the melting point of the resulting mixtures compared with their individual components, confirming the formation of the eutectic solvents. In the ^{1}H NMR spectra, changes were observed in the signal corresponding to the hydrogen attached to the oxygen, with some signals shifting downfield and others disappearing entirely. Similarly, modifications in the alcohol band around 3250 cm^{-1} in the FTIR spectra further corroborated the successful formation of eutectic solvents.

Once the eutectic solvents were formed, they were incorporated into eutectogels by adding acrylic monomers to form polymer networks and subjecting the mixtures to UV polymerization. Four different acrylates were tested as gelators (Figure 1b), including PEGDA (poly(ethylene glycol) diacrylate), DPGDA (dipropylene glycol diacrylate), EGDMA (ethylene glycol dimethacrylate), and HA (ethyl hexyl acrylate). Eutectic solvent-gelator combinations that resulted in homogenous and stable membrane materials were selected for further study. Five eutectogels were prepared with a composition of 60 wt% eutectic solvent and 40 wt% acrylate polymer network, namely P_{4444}Cl-Thy DPGDA, N_{bz444}Cl-Thy with DPGDA, ChCl-Thy with HEMA-EGDMA, C8-Thy with PEGDA, and C8-Men with PEGDA, as shown in Figure 1c.

To evaluate the hydrophobic nature of the eutectogels, surface energy measurements were performed, which revealed the intermolecular forces present at the surface of the eutectogel samples (Figure 2a). The results provide both dispersive and polar components of the materials. The ChCl-Thy HEMA-EGDMA eutectogel exhibited the highest polar component, identifying it as the most hydrophilic material. In contrast, the N_{bz444}Cl-Thy- and C8-Thy-based eutectogels, which combine long hydrocarbon chain hydrogen bond acceptors, demonstrated enhanced surface hydrophobicity. The eutectogels with the lowest polar component, and thus the highest hydrophobic nature, were found to be those based on P_{4444}Cl-Thy and C8-Men.

Although the surface energy results provide valuable insights into the hydrophobic nature of the materials, water uptake measurements are crucial to confirm the behavior of the eutectogels when exposed to water (Figure 2b). The results revealed that eutectogels containing salt-based eutectic solvents (P_{4444}Cl-Thy, Nbz_{444}Cl-Thy, and ChCl-Thy) experienced significant weight loss, ranging from 10 to 20% after immersion in water. The eutectogel based on C8-Thy exhibited a smaller weight loss of 6% relative to its initial weight. In contrast, the menthol-based eutectogel demonstrated a distinct behavior, initially absorbing water and increasing its weight by 15%, followed by a gradual weight loss, eventually stabilizing at 98% of its initial weight. After 48 h, all eutectogels exhibited varying



c) Eutectogels

Figure 2. a) Surface energy (dispersive component - dash fill; polar component - solid fill), b) water uptake, and c) aspect of eutectogels after immersion in water for 48 h.

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degrees of phase separation and noticeable changes in appearance (Figure 2c), which could influence their performance in underwater environments.

To improve the underwater stability of the two eutectogels that demonstrated the best water uptake results and also to minimize phase separation, 10 wt% ethyl hexylacrylate (HA) monomer was mixed with 30 wt% PEGDA. This modification was introduced not only to enhance the hydrophobicity of the polymeric acrylic network but also to increase its compatibility with the eutectic solvent.[26] The effect of incorporating HA into the eutectogel was assessed by analyzing the thermal properties of the resulting materials (Figure 3).

The C8-Thy and C8-Men eutectic solvents exhibited melting temperatures (T_m) of 10 °C and 6 °C, respectively (Figure 3a), and a degradation temperature of approximately 100 °C (Figure 3b). The melting temperatures of the eutectic solvents were lower than those of the individual components: – menthol (T_m= 42 °C), thymol (T_m=49 °C), and octanoic acid T_m = 16 °C); indicating the formation of eutectic solvents. On the other hand, the eutectogels did not show any glass transition down to −60 °C, indicating an even lower glass transition temperature (T_g) value. However, the eutectogels exhibited a less pronounced melting point signal, corresponding to the melting temperature of the eutectic solvents. This can be attributed to a slight excess of the eutectic solvent that could not be entirely entrapped within the polymer network.

TGA indicated that the eutectic solvents underwent a single step of degradation, starting at approximately 115 °C (Figure 3b). For eutectogels based on menthol as the hydrogen bond donor, a two-step degradation process was observed. The first degradation step starts at the same temperature as that of the eutectic solvent, indicating degradation of the eutectic component. The second degradation step corresponds to the polymeric material and occurs at approximately 355 °C. A similar degradation trend was observed for the thymol-based eutectogels, in which the eutectic solvent first degraded, followed by polymer degradation at approximately 360 °C.

The mechanical properties of the eutectogels were evaluated through rheological studies to examine the effects of the eutectic solvent and the addition of HA. In general, the eutectogels displayed typical rubber-like behavior, with the storage modulus (G′) surpassing the loss modulus (G′′) up to 8 % strain, indicating elastic (solid-like) behavior (Figure 4a). The results also revealed that the variation in thymol or menthol in the chemical structure of the eutectic solvent did not significantly affect the storage modulus of the eutectogels, as they both showed a value of approximately 750 Pa. However, for thymol-based eutectogels, the addition of HA notably reduced the storage modulus from 800 to 200 Pa. This decrease can be attributed to the addition of HA, which led to a lower crosslinking density within the eutectogel. In contrast, this effect was not observed for menthol-based membranes, as their G′ values remained almost unchanged.

The frequency sweep analysis showed that the eutectogels predominantly exhibited elastic solid behavior, with G′ > G′′, indicating efficient energy storage rather than dissipation (Figure 4b). The storage modulus increased slightly with frequency, which is attributed to the enhanced resistance of the material to deformation at higher frequencies. This behavior can be explained by the crosslinked network, which restricts the ability of the material to relax and causes it to behave more elastically.

Based on the water uptake results and promising rheological properties, the C8-Men PEGDA-HA eutectogel was selected to evaluate its potential to absorb contaminants of emerging concern (CECs), including diclofenac, iopromide, cefazolin, dichlorophenol, and bisphenol A, from aqueous solutions. Figure 5 illustrates the experimental sorption kinetics, which shows that equilibrium was reached after approximately 5 h of incubation. The C8-Men PEGDA-HA eutectogel membrane exhibited high sorption capacities for bisphenol A, dichlorophenol and diclofenac. Despite that, the sorption of diclofenac in C8-Men PEGDA-HA eutectogel is nearly half the amount of bisphenol A and dichlorophenol. Additionally, this eutectogel

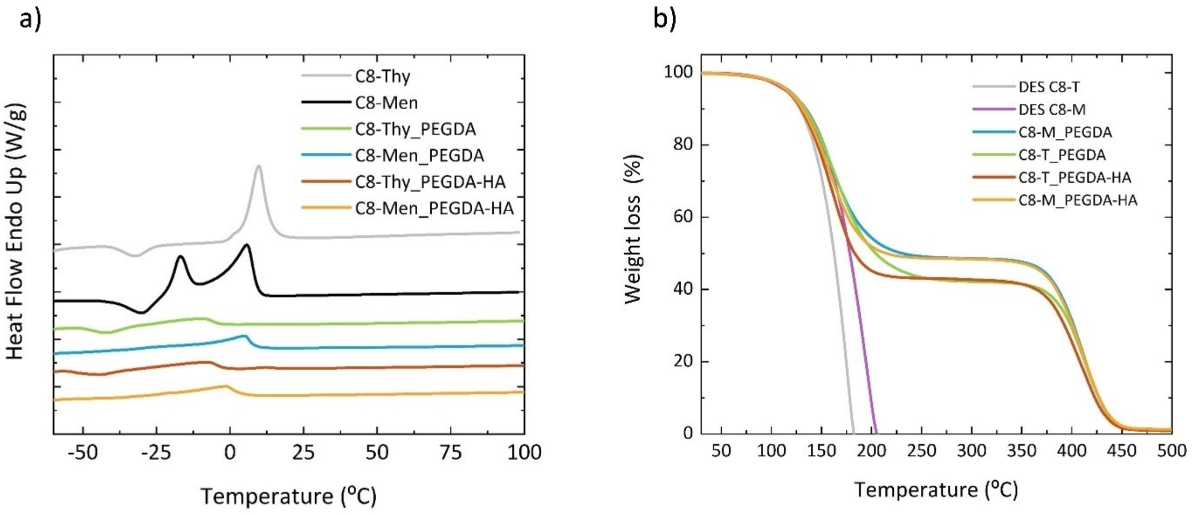

Figure 3. a) DSC and b) TGA analyses of C8-Thy and C8-Men based eutectic solvents and eutectogels.

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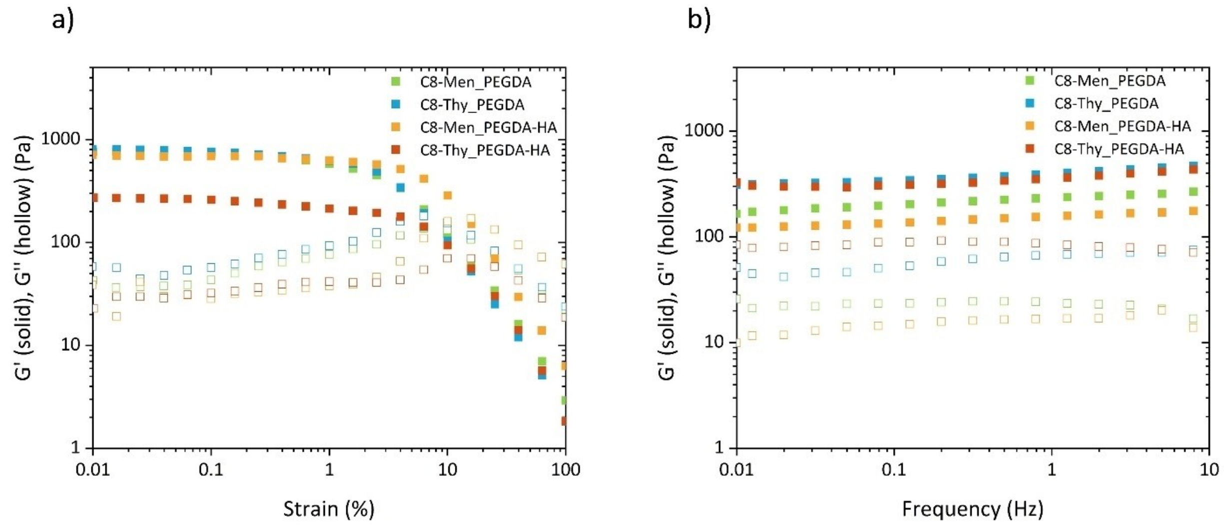

Figure 4. a) Strain sweep and b) frequency sweep of eutectogel membranes composed of C8-Men and C8-Thy eutectic solvents.

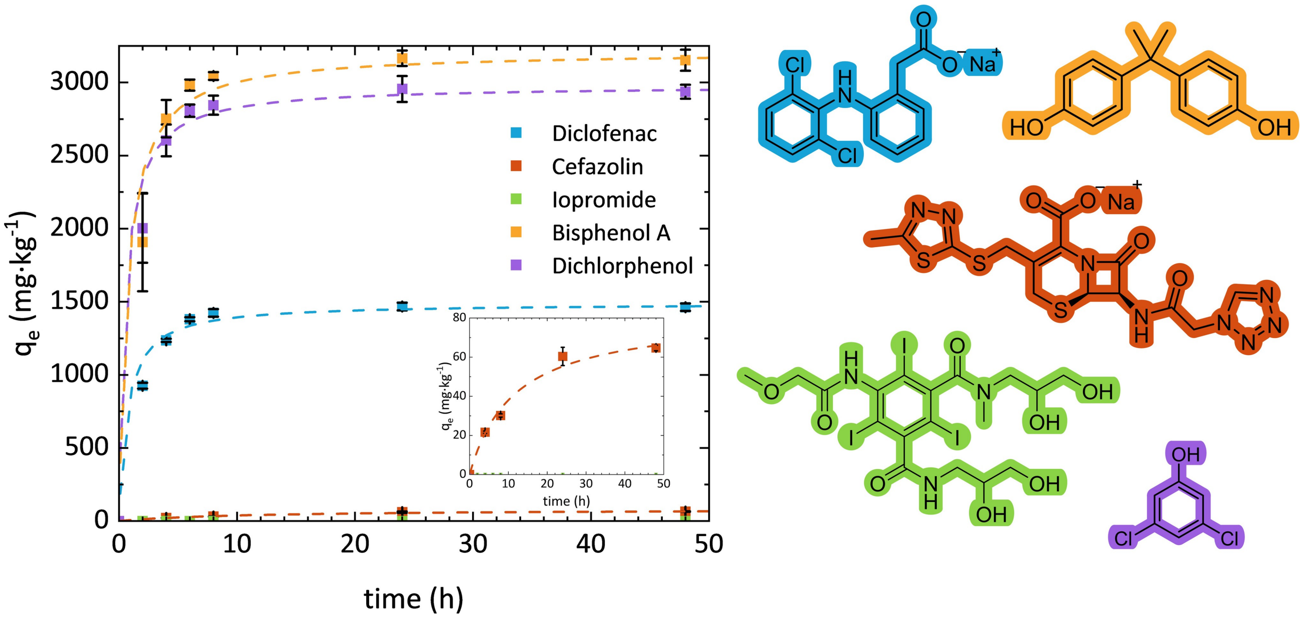

Figure 5. CECs uptake capacity in the menthol-based eutectogel under the pseudo-second-order linear model as a function of time.

was ineffective at adsorbing iopromide and exhibited a very low sorption of cefazolin.

The pseudo-second-order kinetic model (equation 1) was employed to fit the experimental data, as follows:

$$\frac{t}{q_t} = \frac{1}{kq_e^2} + \frac{1}{q_e} \quad (1)$$

where t represents time (hours), q_t and q_e denote the amount of CECs adsorbed (mg·kg^{-1}) by C8-Men PEGDA-HA eutectogel at time t and at equilibrium, respectively, and k is the pseudo-second-order rate constant (kg·mg^{-1}·h^{-1}). The fitting results for the pseudo-second-order linear model are illustrated in Figure 5 and summarized in Table 1. The C8-Men PEGDA-HA eutectogel demonstrated remarkable sorption capacity, absorbing approximately 3000 mg·kg^{-1} of bisphenol A and dichlorophenol at equilibrium. In the case of diclofenac, the sorption capacity was nearly 1500 mg·kg^{-1}, while for cefazolin, it is 80 mg·kg^{-1}. These results suggest that the eutectogel exhibits steric hindrance, limiting its ability to sorb molecules of larger dimensions, such

Table 1. Kinetic parameters of the pseudo-second-order model for CECs uptake using the C8-Men PEGDA-HA eutectogel.

	R^2	q_e (mg·kg^{-1})	k_2 (kg·mg^{-1}·h^{-1})
Diclofenac	0.999	1490.0	0.0010
Cefazolin	0.987	81.7	0.0010
Bisphenol A	0.999	3213.0	0.0004
Dichlorphenol	0.999	2981.1	0.0006

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as iopromide and cefazolin. This behavior can be influenced by the dense morphology of the eutectogel (Figure S15), which may restrict the accessibility of larger molecules to the sorbent material.

Few studies have explored the use of eutectic solvent-based materials for water purification. For instance, supported liquid membranes incorporating a eutectic solvent as the liquid phase have been developed by embedding a decanoic acid:octanoic acid eutectic mixture into an inert PVDF porous membrane. These membranes were tested for the removal of diclofenac and bisphenol A from water. Although they exhibited a higher sorption capacity for diclofenac, their performance for bisphenol A was lower than that of the eutectogel tested in this study.[30] Beyond pharmaceutical contaminants, eutectogels have also demonstrated effectiveness in removing dye pollutants. In particular, L-phenylalanine-based eutectogels demonstrated high sorption capacities of approximately $1.93 \cdot 10^6$ $mg \cdot kg^{-1}$ for rhodamine B.[29] On the other hand, supramolecular iongels, composed of the [p-C_{12}][Mal] salt and the [bmim][PF_6] ionic liquid, have also been investigated for the removal of rhodamine B and methyl orange dyes from water, with sorption capacities of about 800 $mg \cdot kg^{-1}$ and 600 $mg \cdot kg^{-1}$, respectively.[31] Furthermore, catechol-based poly(ionic liquid) hydrogels have shown promising results in dye removal, exhibiting sorption capacities of around 500 $mg \cdot kg^{-1}$ for Eosin Y and 250 $mg \cdot kg^{-1}$ for methylene blue.[32]

Although the above-mentioned studies reported sorption results that align with our findings, a direct comparison is not feasible owing to the distinct nature of the materials tested. Differences in the composition, structural characteristics, and types of contaminants tested further complicate these comparisons. Each material possesses unique properties that govern specific interactions with contaminants, resulting in variations in sorption capacities. Thus, the results obtained in this study highlight both the novelty and potential of polymer network hydrophobic eutectogels for advancing sorption-based water treatment technologies.

Conclusions

A set of hydrophobic eutectogels were developed aiming at the removal of contaminants of emerging concern from water. The results demonstrated that thymol- and menthol-based eutectogels exhibited differing hydrophobicity and water uptake behavior, with the menthol-based eutectogel outperforming the others in terms of stability and capacity to repel water. The incorporation of ethyl hexylacrylate (HA) enhanced the hydrophobicity and compatibility of the PEGDA network with the eutectic solvent, significantly improving the overall performance. Rheological studies confirmed the rubber-like behavior of the eutectogels, with menthol-based variants exhibiting a higher storage modulus and better mechanical properties.

Sorption experiments revealed that the menthol-based eutectogel effectively absorbed bisphenol A and dichlorophenol, demonstrating high sorption capacities, closely following diclofenac. However, minimal sorption of iopromide and cefazolin was observed. A pseudo-second-order kinetic model was employed to fit the experimental data, indicating that equilibrium was reached within approximately five hours of incubation.

This study highlights the versatility of hydrophobic eutectogels for water purification and demonstrates their potential as efficient adsorbents for various water contaminants. Future studies should further explore the mechanisms driving sorption and investigate methods to optimize the performance of hydrophobic eutectogel materials across different pollutant types.

Supporting Information

Electronic supplementary information (ESI) available: Differential Scanning Calorimetry, Proton Nuclear Magnetic Resonance and Fourier Transport Infrared spectroscopy results. Calibration lines of contaminant concentration versus absorbance by UV-Vis absorption. UV-Vis absorbance curves at different times.

Data Availability

The data that support the findings of this study are openly available in MAR2PROTECT Zenodo Repository at https://doi.org/10.5281/zenodo.15014308

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Conflict of Interests

The authors declare no conflict of interest.

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Data Availability Statement

The data that support the findings of this study are openly available in Zenodo at https://doi.org/10.5281/zenodo.15014308, reference number 0.

Keywords: hydrophobic eutectic solvents · eutectogels · contaminants of emerging concern · water treatment · sorption

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