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Organogels based on aromatic amino acids with appended long-chain fatty acids for sustainable oil spill recovery and environmental remediation

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Various organogelator molecules based on fatty-acid-appended amino acids have been synthesized *via* amide coupling between an aromatic amino acid (L-phenylglycine/L-phenylalanine) and fatty acid (palmitic acid/lauric acid), and their organogelation abilities have been tested in various organic solvents, including simple hydrocarbons, aromatic solvents, and oils, namely, kerosene and diesel. Palmitic-acid-appended L-phenylalanine (**C_{15}-Phe**) and L-phenylglycine (**C_{15}-Phg**) undergo stable organogelation in organic solvents including iso-octane, heptane, hexane, cyclohexane, methylcyclohexane, petroleum ether, kerosene and diesel. However, lauric-acid-appended L-phenylglycine (**C_{11}-Phg**) and L-phenylalanine (**C_{11}-Phe**) undergo transient organogelation and clear solution formation under the same conditions, respectively. Interestingly, the minimum gelation concentration of the organogelator **C_{15}-Phg** has been estimated to be lower than that of the organogelator **C_{15}-Phe**. Moreover, **C_{15}-Phg** organogels in all hydrocarbon solvents show a helical nanofibrillar morphology; however, **C_{15}-Phe** shows a nanobelt-like straight fibrillar morphology, except in isooctane. **C_{11}-Phg** also shows a helical nanofibrillar ribbon-like morphology. Powder XRD and FT-IR studies revealed the involvement of π–π stacking, hydrogen bonding and van der Waals interactions in their gel states. The organogels of **C_{15}-Phg** in diesel and kerosene show thixotropic behaviour. The thermal stabilities of the **C_{15}-Phg** organogels have been found to be greater than those of **C_{15}-Phe**. A phase-selective gelation study has been performed in a diesel–water mixture for **C_{15}-Phg**, in which the gel layer could be easily separated to recover the oils, demonstrating the promising applicability of **C_{15}-Phg** in oil spill recovery from marine and aquatic water. This study showcases the green and sustainable development of oil spill recovery and environmental remediation by using simple chemistry to save marine and aquatic life.

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Environmental significance

The formation of supramolecular organogels based on fatty-acid-appended amino acids in kerosene and diesel demonstrates oil spill recovery using phase-selective gelation through environmentally friendly, green and sustainable pathways. Such biomolecule-based environmental remediation is promising for the protection of the environment from immediate and long-term damage, which is the current state of the art in research. This approach enables the reduction of marine, aquatic and air pollution by balancing the biodiversity in water, marine air, soil, and the food chain.

1 Introduction

Low-molecular-weight organogelators are recognized as versatile advanced smart functional materials due to their impactful applications in different fields, including oil spill recovery,[1–8] toxic dye and metal ion removal,[9–14] drug release,[15,16] anti-freezing agents,[17] health monitoring materials,[18] photonic switches,[19] dental composite carriers[20] and dye-sensitized solar cells.[21] In recent years, an alarming situation has arisen due to oil spills in seawater, which have led to serious damage to marine life and the destruction of sensitive habitats such as coral reefs and mangroves. Oil spills, which occur mainly in seawater, cause immediate and long-term environmental damage by physically smothering wildlife and poisoning animals through toxic exposure. They also affect biodiversity by contaminating the marine water, air, and soil and disrupting food chains. The release of volatile organic compounds (VOCs) and particulate matter into the atmosphere also occurs through evaporation and controlled burning processes during oil clean-up; these VOCs contribute to smog, which poses acute

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respiratory risks to wildlife and humans. Unless green, sustainable, and successful oil spill recovery processes and techniques can be developed, oil spills present potential long-term effects on populations and ecosystems that can take years to recover. Various methods have been developed to remove oil spills from seawater, including oil skimmers,[22] solidifiers,[23] microbial degradation,[24] and sorbents.[25] However, their large-scale practical use is still under consideration due to the long process times, the toxicity of the materials, and the high cost of the processes. Many researchers have dedicated their efforts to the recovery of spilled oil from seawater *via* phase-selective gelation techniques. Here, organogelator molecules assemble into three-dimensional networks, entrapping huge quantities of organic solvents/oils through weak non-covalent interactions, including π–π stacking, van der Waals, and hydrogen-bonding interactions. As these non-covalent interactions are weak, the structural integrity of the gelator molecules remains unaltered. Thus, after collection, the gel in the oil can be recovered easily by distillation, and the gelator can be reused in most cases. In the year 2001, Bhattacharya and his co-workers first reported phase-selective gelation with a fatty-acid-appended amino acid gelator, which effectively formed an organogel in various hydrocarbon solvents as well as oils, including kerosene, petrol and paraffin oil.[26] Since then, several different types of gelator molecules that can remove oils from water through gel formation have been developed.[27–37] Various sugar-based molecules,[27–31] amino-acid derivatives and peptides,[32–39] asymmetric amino acids and aliphatic-chain-functionalized naphthalene diimide,[40] cellulose nanofibrils,[41] urea,[42] lime juice with long-chain amines,[43] imines,[44] vanillin,[45] and other molecules[46] have evolved over time for oil spill recovery. Previously, Banerjee and co-workers worked on myristic-acid-appended amino-acid-based gelator molecules.[35] They showed that a myristic-acid-appended phenylglycine-based gelator molecule can effectively form an organogel in various organic solvents, including kerosene and diesel and its oil spill recovery.[35] Vujičić and co-workers prepared four (*N*-alkyloxalamido)-amino-acid amide-based gelator molecules, one of which showed very high gelation capabilities in diesel (MGC = 0.015) and petrol (MGC = 0.012).[47] Instant phase-selective gelation has been achieved at room temperature in a diesel–water mixture.[47] Frostad's group prepared a long-chain unsaturated-fatty-acid-appended amino-acid-based gellant that can form a macroscopic structure in both aqueous and organic solvents.[48] The macroscopic structure that formed in the aqueous layer was able to absorb oil slowly, replacing water; no other solvent was required for the deployment of the gelator molecules in this oil spill recovery process.[48] Gupta's group synthesised a list of *N*-hydroxy alkanamides that can form organogels in a variety of plant-based lipophilic solvents. They also studied the use of these types of gelators to remove Cu(II) and Fe(II) metal ions from water through chelation interaction.[49] Yadav's group prepared a D-/L-arabinose-modified organogelator that can form a gel in various organic solvents and can take part in a photochemical reaction in the gel state upon irradiation at a particular wavelength.[50] De's group prepared polymer-appended amino-acid-based gelator molecules that can form a gel in some organic solvents, as well as in diesel.[51] Molla's group prepared an azobenzene-containing photo-switchable organogelator that can form a gel *via* *J*-type aggregation in toluene and also *H*-type aggregation in methylcyclohexane.[52] It can be used for the recovery of oil from diesel/water and petrol/water mixtures *via* organogelation.[52] Sureshan and co-workers prepared modified D-glucose organogelators that can be used directly on the seawater surface without applying any gelation triggers for the phase-selective gelation in oil spills on seawater.[53] Roy's group prepared various amino-acid-appended, cholesterol-based gelator molecules capable of absorbing toxic organic dyes from water that can also remove oil from water through a phase-selective organogelation process.[54] However, to overcome the issues of high synthesis costs, lack of cheaper starting materials, high MGC (minimum gelation concentration) values, slow gel-forming kinetics, and non-thixotropic natures, the design and development of novel amino-acid-based organogelators remains important. In this context, we have materialized organogels by synthesizing four new amino-acid-based long-chain fatty acids appended amides that are thixotropic, mechanoresponsive, and low-cost, with easy synthetic steps, reproducible, lower MGCs, and relatively faster gel formation kinetics. In our research, we have used two fatty acids: one with a greater number of carbons in its aliphatic chain (palmitic acid), and one has fewer carbons (lauric acid), compared to the previously used fatty acid (myristic acid)[35] and coupled with the same amino acids (L-phenylalanine and L-phenylglycine), to prepare a total of four long-chain fatty acid–amino acid conjugates for investigating the roles of long-chain fatty acids and amino acids in self-assembly and gelation. We have synthesized and purified four new compounds, **C$_{11}$-Phg**, **C$_{11}$-Phe**, **C$_{15}$-Phg**, and **C$_{15}$-Phe** (Fig. 1). It has been found that **C$_{15}$-Phg** and **C$_{15}$-Phe** form organogels selectively in hydrocarbon-based organic solvents, including kerosene and diesel oils, whereas **C$_{11}$-Phg** forms transient gels that precipitate after 1 to 2 days, and **C$_{11}$-Phe** fails to form gels. **C$_{15}$-Phg** has been found to be the superior gelator due to its lower MGCs and higher thermal stability compared to **C$_{15}$-Phe** organogels. Interestingly, in kerosene and diesel, **C$_{15}$-Phg** forms thixotropic gels. The selective thixotropic organogelation in kerosene and diesel is promising for real-world usage, as after the breakage of the gels, the self-assembled viscous solutions of the gels have the ability to reform without losing the recovered oils from the marine aquatic environment. To the best of our knowledge of the literature, most of the organogelators applied for oil spill

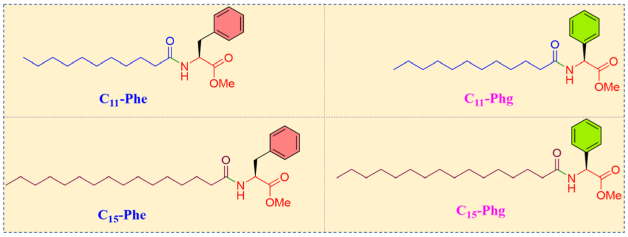

Fig. 1 Synthesized fatty-acid-appended amino acid derivatives chemical structures.

recovery are not thixotropic in nature or not mentioned about their thixotropic behaviour. Moreover, palmitic-acid-appended L-phenylglycine shows helical-ribbon-like nanostructures; however, palmitic-acid-appended L-phenylalanine forms a nanobelt-like straight fibrillar morphology. A previous study by Banerjee's group[35] and this study suggest that L-phenylglycine induces a chiral helical nanofibrillar morphology. A phase-selective gelation study has been performed in a diesel–water mixture for **C_{15}-Phg**, from which the gel layer can easily be separated to recover oils and prevent oil spills from causing aquatic marine pollution. This research describes the roles of different fatty-acid attached amino-acid-appended gelators in phase-selective organogelation and their application in oil spill recovery and environmental remediation, following green and sustainable platforms.

2 Experimental section

2.1 Materials and methods

2.1.1 Materials. The amino acids L-phenylalanine and L-phenylglycine, and the long-chain fatty acids lauric acid, palmitic acid, HOBt, disodium hydrogen phosphate, dihydrogen sodium phosphate, sodium carbonate, sodium chloride, all the organic solvents used in this work, and 100–200 mesh size silica were purchased from Sisco Research Laboratories Pvt. Ltd. (SRL)–India. DCC was purchased from Molychem India LLP. Hydrochloric acid was purchased from Avra Synthesis Pvt. Ltd. Ethyl acetate and petroleum ether were purchased from Rankem Laboratory.

2.1.2 Syntheses of C_{11}-Phe, C_{11}-Phg, C_{15}-Phe, and C_{15}-Phg. The syntheses of all these gelators (Fig. 1) were performed by following a solution-phase peptide synthetic protocol using the coupling reagent DCC with the additive HOBt [Scheme S1]. After synthesis, the gelator molecules were purified by column chromatography using 100–200 mesh size silica gel with an ethyl acetate and petroleum ether solvent mixture as the eluent. All four purified compounds (**C_{11}-Phg**, **C_{11}-Phe**, **C_{15}-Phg**, and **C_{15}-Phe**) appeared as powdered white solids. The purified compounds were used for the organogelations and all other studies. All these compounds were thoroughly characterised by mass spectrometry, elemental and NMR analyses (see synthetic protocols and Fig. S1 to S16, SI). A detailed experimental section, sample preparation, instrumentation, and methods of analysis are provided in the SI.

3 Results and discussion

3.1 Gelation studies and gel melting point measurement

We studied the gelation abilities and thermal stabilities of four gelator molecules, **C_{11}-Phe**, **C_{11}-Phg**, **C_{15}-Phe**, and **C_{15}-Phg** (Fig. 1). Two different long-chain fatty acids, with 11 or 15 methylene carbon atoms, excluding the carboxylic acid carbon, were judiciously coupled with two different amino acid (L-phenylalanine and L-phenylglycine) methyl esters to understand the effects of alkyl chain length and aromatic amino acids in organogelation, keeping the aromatic amino acids the same as in a previous study.[35]

20 mg of the gelator molecules were taken into 3 mL glass vials, and then 1 mL of various organic solvents, including diesel and kerosene-like petroleum oils, were added separately to the glass vials. The vials were heated using a hot air gun with continuous shaking by hand until all the gelator molecules had dissolved, followed by cooling in ice-cold water to obtain stable self-supported organogels (Fig. 2). The inverted vial test method was used to confirm organogelation. Sometimes, it was observed that after being kept at room temperature after gelation, the gel reverted to a solution state, and interestingly, stable gel formation occurred again with time. This procedure was followed for the gelation and self-assembly of the synthesized compounds **C_{11}-Phe**, **C_{11}-Phg**, **C_{15}-Phe**, and **C_{15}-Phg**.

Various organic solvents, namely, isooctane, heptane, cyclohexane, methylcyclohexane, hexane, petroleum ether, kerosene, diesel, 1,2-dichlorobenzene, benzene, toluene, chlorobenzene, isopropyl alcohol, and *n*-hexanol, were selected for the gelation studies. Among the four gelator molecules **C_{11}-Phe**, **C_{11}-Phg**, **C_{15}-Phe**, and **C_{15}-Phg**, it was observed that **C_{15}-Phg** showed superior gelation ability (Tables 1 and 2), as well as the highest thermal stability in many of these different hydrocarbon solvents, including isooctane, heptane, cyclohexane, methylcyclohexane, hexane, petroleum ether, kerosene, and diesel. The gelator **C_{15}-Phe** forms stable gels (Fig. S17) in these solvents; however, their MGCs are higher, and their thermal

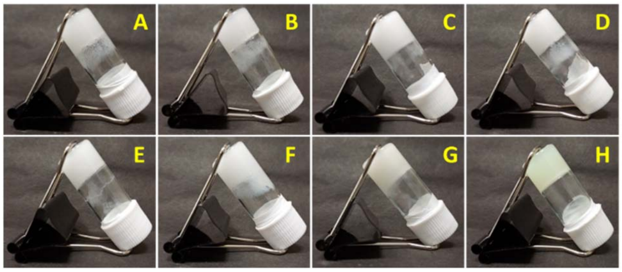

Fig. 2 Organogels of **C_{15}-Phg** in isooctane (A), heptane (B), cyclohexane (C), methylcyclohexane (D), hexane (E), petroleum ether (F), kerosene (G), and diesel (H).

Table 1 Outcomes of the gelation experiments of **C_{11}-Phe**, **C_{11}-Phg**, **C_{15}-Phe**, and **C_{15}-Phg** in different organic solvents

Solvent	C_{11}-Phe	C_{11}-Phg	C_{15}-Phe	C_{15}-Phg
Isooctane	P	MS	G	G
Heptane	P	MS	G	G
Cyclohexane	S	MS	G	G
Methylcyclohexane	S	MS	G	G
Hexane	S	MS	G	G
Pet ether	S	MS	G	G
Kerosene	S	MS	G	G
Diesel	S	MS	G	G
1,2-Dichlorobenzene	S	S	S	S
Benzene	S	S	S	S
Toluene	S	S	S	S
Chlorobenzene	S	S	S	S
IPA	S	S	S	S
n-hexanol	S	S	S	S

Table 2 MGCs of C_{15}-**Phe** and C_{15}-**Phg** in different organic solvents, and the gel melting point temperatures at concentrations higher than their MGCs

		C_{15}-Phg		C_{15}-Phe	
Solvent	Concentration at which T_{gel} was measured (% w/v)	MGCs (% w/v)	T_{gel} (°C)	MGCs (% w/v)	T_{gel} (°C)
Isooctane	1.1	0.4	46.66 ± 0.577	0.6	40 ± 1.00
Heptane	1.0	0.4	41.66 ± 1.154	0.6	35.66 ± 0.577
Cyclohexane	2.3	1.18	36.33 ± 1.154	1.9	31 ± 1.00
Methylcyclohexane	2.4	0.84	42 ± 0.00	1.6	35 ± 0.00
Hexane	1.1	0.37	42.33 ± 0.577	0.7	35.66 ± 0.577
Pet ether	1.3	0.56	42 ± 1.00	0.9	35 ± 1.00
Kerosene	2.4	0.95	46 ± 0.00	2	34.66 ± 0.577
Diesel	1.2	0.59	42.66 ± 0.577	1	34.66 ± 0.577

stabilities are lower compared to those of C_{15}-**Phg** (Table 2). C_{11}-**Phg** formed gels in these solvents; however, after 1–2 days, the gels had disintegrated, and precipitation had formed (Fig. S18). Interestingly, C_{11}-**Phe** did not form a gel in any of these solvents; instead, the formation of precipitation and transparent solutions were observed (Fig. S19). The outcomes of the gelation experiments are given in tabular form (Table 1). The minimum gelation concentrations and thermal stabilities of these organogels have been tabulated in Table 2. Nevertheless, it was observed that after heating to dissolve the gelator molecules, immediate cooling in ice-cold water resulted in better gel formation compared to that obtained without cooling.

To investigate their thermal stability, the melting point temperatures of the gels prepared in glass vials were determined by dipping the vials into a digitally temperature-controlled water bath for 5 minutes and testing the stability of the gels using the vial inversion method. Every 5 minutes, the temperature was increased by 1 °C, and the gel melting temperatures were recorded. T_{gel} measurements for all the organogels were performed in triplicate for each concentration of each gelator in the different solvent systems, and their standard deviation errors are given in Table 2. As the gelator molecules C_{15}-**Phg** and C_{15}-**Phe** form stable gels, the gel melting temperatures for these two organogelator molecules were determined in various solvents, while keeping the concentration constant for both gelator molecules in each solvent. The melting temperatures of the organogels in isooctane, heptane, cyclohexane, methylcyclohexane, hexane, petroleum ether, and kerosene solvents are given in Table 2. The concentration-dependent gel melting temperatures of C_{15}-**Phg** and C_{15}-**Phe** in diesel at four different concentrations are shown in Fig. 3.

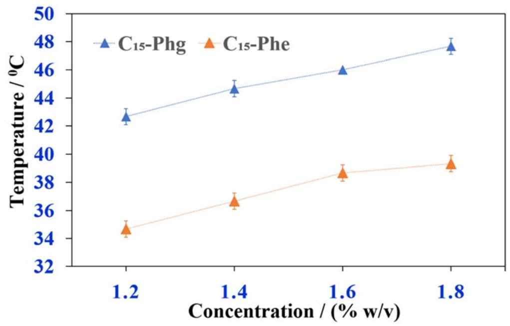

Fig. 3 Gel melting temperatures of C_{15}-**Phg** and C_{15}-**Phe** gels in diesel at various concentrations.

3.2 Rheological measurement

The viscoelastic properties of the gels were measured *via* rheological measurements. Gels of C_{15}-**Phg** and C_{15}-**Phe** in diesel and kerosene at 1.4% (w/v) and 2.4% (w/v), respectively, were used for the angular frequency sweep experiment. As shown in Fig. 4, the results suggest a higher storage modulus (G′) compared to the loss modulus (G″) under a constant shear stress of 0.1%. This indicates stable organogelation [Fig. 4 and S20]. Interestingly, in kerosene and diesel, thixotropic organogelation was observed, as is evident from Fig. 4 and S20.

3.3 Morphological study

To understand their morphological behaviour, field-emission scanning electron microscopy (FE-SEM) was performed for C_{11}-**Phe**, C_{11}-**Phg**, C_{15}-**Phe**, and C_{15}-**Phg** in various organic solvents. The C_{15}-**Phg** gelator molecules in their gel state exhibit right-handed helical fibres in isooctane, heptane, cyclohexane,

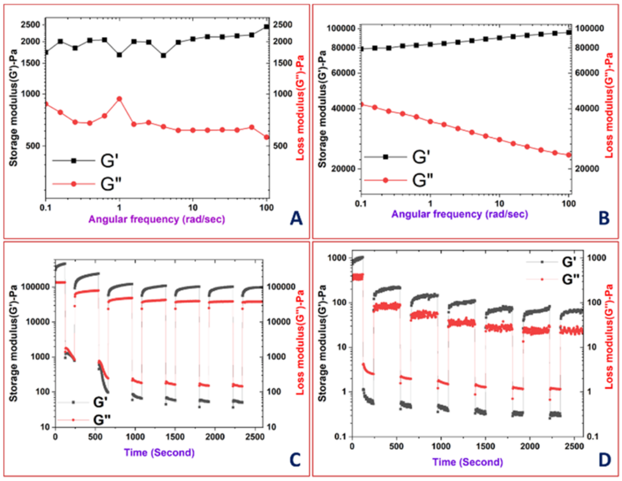

Fig. 4 Frequency sweep profiles for the gels of C_{15}-**Phg** in kerosene (A) and diesel (B). Thixotropic profiles of gel breaking and recovery of the gels of C_{15}-**Phg** in kerosene (C) and diesel (D).

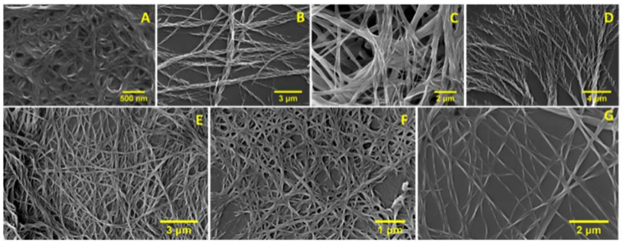

Fig. 5 FE-SEM images of the organogels of the gelator **C_{15}-Phg** in isooctane (A), heptane (B), cyclohexane (C), methylcyclohexane (D), hexane (E), petroleum ether (F) and kerosene (G).

methylcyclohexane, hexane, petroleum ether, and kerosene solvents [Fig. 5]. The **C_{15}-Phe** gelator molecules in their gel state show right-handed twisted fibres in isooctane, whereas in other solvents, they show straight nanobelt-type fibres [Fig. S21]. Interestingly, **C_{11}-Phg** shows twisted fibres in isooctane [Fig. S21] and kerosene [Fig. S22]. FE-SEM analysis of the **C_{11}-Phe** solution in kerosene is shown in Fig. S22.

3.4 FT-IR analysis

Fourier-transform infrared (FT-IR) analysis was performed for all the organogels obtained from **C_{15}-Phg** and **C_{15}-Phe** in their xerogel states in the solvents isooctane, heptane, cyclohexane, methylcyclohexane, hexane, petroleum ether, kerosene, and diesel. FT-IR analysis was also carried out for the amorphous solid compounds to understand the involvement of different functional groups in various non-covalent interactions (Fig. S23). The pure amorphous compound **C_{15}-Phg** shows characteristic peaks (Fig. 6) at 3309.84 cm^{-1} for N–H stretching, 2914.44 cm^{-1} and 2848.86 cm^{-1} for the alkyl C–H stretching, 1735.93 cm^{-1} for the C=O stretching of the ester bond, 1651.06 cm^{-1} for the C=O stretching of the amide bond, and 1529.55 cm^{-1} for the N–H bending/stretching of the amide bond. The amorphous compound **C_{15}-Phe** shows corresponding peaks at 3338.78 cm^{-1}, 2914.44 cm^{-1}, 2848.86 cm^{-1}, 1751.36 cm^{-1}, 1647.20 cm^{-1}, and 1525.69 cm^{-1} (Fig. S24). The xerogels of **C_{15}-Phg** obtained in different solvents show a small increase in the N–H stretching frequencies in all the solvents, suggesting the involvement of hydrogen-bonding interactions,[9] except in the case of cyclohexane, in which no change was observed compared to the amorphous state. The C=O stretching frequency of the ester bond was red-shifted for the solvents methylcyclohexane and diesel; for the other solvents, no change was observed compared to the amorphous state. At the same time, a blue-shift of the amide C=O stretch was identified for all the solvents except isooctane, in which no change was observed.

The N–H bending frequency was red-shifted for the organogels in the solvents isooctane, cyclohexane, petroleum ether, and kerosene, whereas for diesel it was shifted to the blue (Fig. 6 and Table S1), suggesting the involvement of hydrogen-bonding interactions.[9] The xerogels obtained from **C_{15}-Phe** show blue-shifts of the N–H stretching frequency in all these solvents (Fig. S24).

The C=O stretching frequency of the ester bond remains unchanged for the native compound and for the xerogels obtained from the corresponding organogels in all the solvents except diesel, for which a blue-shift was observed. The N–H bending vibration was red-shifted for the xerogels derived from the isooctane, petroleum ether, and diesel organogels, whereas for the other xerogels, no change was observed compared to the amorphous solid. No change was observed in the alkyl C–H stretching frequencies (Fig. S23, S24 and Table S2).

3.5 X-ray diffraction study

X-ray diffraction studies were performed for the xerogels obtained from the organogels in different solvents to identify possible non-covalent interactions involved in the formation of the organogels. All the xerogels obtained from **C_{15}-Phg** show characteristic peaks between $2\theta = 10.98°$ to $2\theta = 11.30°$ (7.82 Å to 8.05 Å), representing the distance between two adjacent β-strands. The peaks found between $2\theta = 22.61°$ to $2\theta = 23.65°$

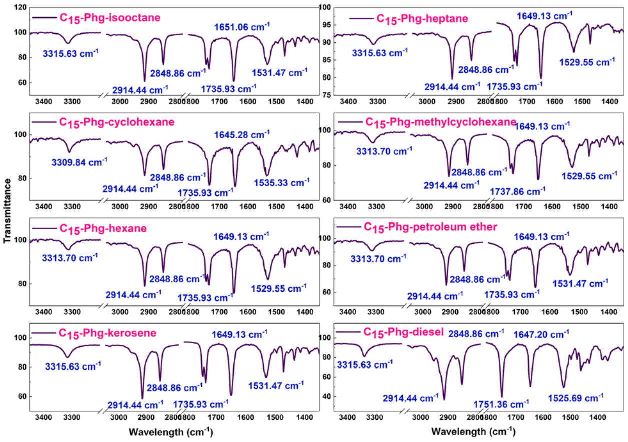

Fig. 6 FT-IR spectra of the xerogels of the gelator **C_{15}-Phg** in isooctane, heptane, cyclohexane, methylcyclohexane, hexane, petroleum ether, and kerosene.

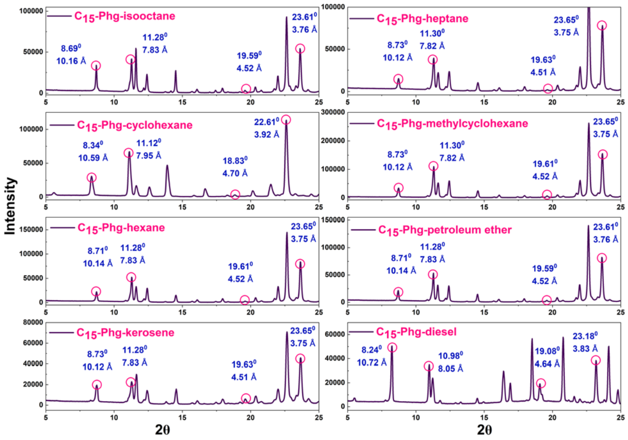

Fig. 7 XRD patterns of the xerogels obtained from the gelator **C_{15}-Phg** in isooctane, heptane, cyclohexane, methylcyclohexane, hexane, petroleum ether, and kerosene solvents.

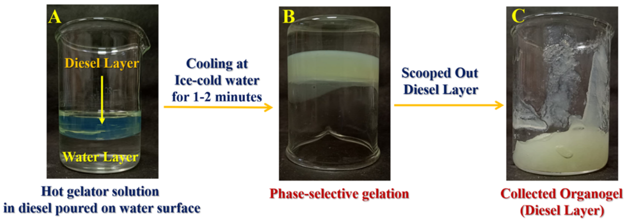

Fig. 8 Immiscible layer formation of water and the C_{15}-Phg gelator hot solution in diesel (A), phase-selective gel formation (B), and separated gel in the diesel layer (C).

(3.75 Å to 3.92 Å) represent π–π stacking interactions that take place through the aromatic rings of the amino acids.[9,35] Another set of peaks were found in between $2\theta = 18.83°$ to $2\theta = 19.63°$ (4.51 Å to 4.70 Å), which represents the hydrogen-bonding distance between two adjacent β-strands (Fig. 7).[9,35] The xerogels obtained from **C_{15}-Phe** also show this type of peak patterns in their XRD profiles, with peaks at $2\theta = 11.04°$ to $2\theta = 11.32°$ (7.81 Å to 8.00 Å), $2\theta = 22.61°$ to $2\theta = 24.16°$ (3.68 Å to 3.92 Å) and $2\theta = 18.49°$ to $2\theta = 20.06°$ (4.42 Å to 4.79 Å) (Fig. S25).[9,35]

3.6 Oil spill recovery

C_{15}-Phg and **C_{15}-Phe** successfully formed organogels in various hydrocarbons and oils selectively. Between these two organogelators, **C_{15}-Phg** was found to be a better gelator molecule. Phase-selective gelation was performed using the **C_{15}-Phg** gelator in diesel. The oil spill recovery experiment was carried out by taking 60 mg of the gelator molecule **C_{15}-Phg** in a glass vial and dissolving it in 5 mL of diesel (at 1.2% w/v, which is twice the MGC of **C_{15}-Phg** in diesel) *via* heating with a hot air gun. This clear, transparent solution was poured into a beaker containing 7 mL of tap water, which was then cooled in ice-cold water for 1–2 minutes to form phase-selective gelation in the diesel layer, as shown by the inverted beaker method (Fig. 8). Later, the gel was scooped out, and the diesel oil was successfully extracted through a distillation setup. This oil spill recovery experiment was also performed using kerosene as a solvent. However, we found that the oil spill experiment involving the gelation of **C_{15}-Phg** in diesel resulted in better gel formation compared to that of **C_{15}-Phg** in kerosene.

4 Conclusions

Herein, we report the gelation abilities of the fatty-acid-appended amino-acid derivatives **C_{15}-Phg**, **C_{15}-Phe**, **C_{11}-Phg**, and **C_{11}-Phe** in various organic solvents and oils. It has been found that the use of phenylglycine substitutes in place of phenylalanine, while keeping the fatty acid residue the same, bestows superior gelation ability. Moreover, the use of the fatty acid with a greater alkyl chain length results in improved gelation ability when keeping the amino acid residue the same. Palmitic-acid-appended phenylglycine ester (**C_{15}-Phg**) is the best gelator among the four compounds (**C_{15}-Phg**, **C_{15}-Phe**, **C_{11}-Phg**, and **C_{11}-Phe**) synthesized, and we studied its selective gelation in various hydrocarbon solvents, including diesel and kerosene oils. L-phenylglycine is an important amino acid that, when appended with long-chain fatty acids, induces the formation of selectively twisting ribbon-like right-handed helical chiral fibres. This study shows the importance of the judicious selection of amino acid side chains and indeed the elegant beauty of the self-assembly in the formation of reproducible chiral right-handed helical nanofibers followed by organogelation. Phase-selective gelation of the gelator **C_{15}-Phg** in a diesel–water mixture, followed by the recovery of the gel layer in diesel from water, represents oil spill recovery. The excellent thixotropic behaviour of the **C_{15}-Phg** organogels in diesel and kerosene is promising for their real-world applicability in oil spill recovery while balancing the biodiversity of marine water, air, soil, and food chains, thus protecting the environment from immediate and long-term damage. This study showcases the green and sustainable development of oil spill recovery and environmental remediation materials using simple chemistry to save marine and aquatic life. In the future, chain length variation of the fatty acids while keeping L-phenylglycine as the fixed amino acid could be attempted to generalize the helical tape formation, self-assembly mechanism, and environmental remediation.

Author contributions

All authors have approved the final version of the manuscript. A. B. was responsible for the syntheses, purifications, organogelations, methodology, investigations, characterizations, data curation, graph preparation from raw data and their calculations, writing the early-stage draft, and formatting. N. J. was responsible for rheological measurements and rheological data curation. B. D. responsible for HR-MS and elemental analyses. S. R. was responsible for project administration, funding acquisition, conceptualization, supervision, writing of the manuscript, review, editing, and finalization of the manuscript and SI.

Conflicts of interest

The authors declare that there are no conflicts to declare.

Data availability

The data supporting this article have been included as part of the supplementary information (SI). Supplementary information: all synthesized compounds were characterised by mass spectrometric, elemental and NMR spectral analyses (Synthetic protocol section and Fig. S1 to S16, SI file). SI file also contains instrumentation techniques and method of analysis, synthetic protocols, gels and solution photographs, FE-SEM, rheology, FT-IR profiles and tables. See DOI: https://doi.org/10.1039/d5va00467e.

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