

Nanomagnetic Organogel Based on Dodecyl Methacrylate for Absorption and Removal of Organic Solvents

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Abstract A novel nanomagnetic organogel was synthesized by *in situ* emulsion polymerization-crosslinking method using dodecyl methacrylate (DDMA) and styrene (St) as monomers, divinylbenzene (DVB) as a crosslinking agent, azobisisobutyronitrile (AIBN) as an initiator, and Fe₃O₄ as a nanomagnetic particle. Modification of the network was carried out by inclusion of the multi-walled carbon nanotubes (MWCNT) into the organogel matrix. The structure of the nanocomposite was characterized using FTIR spectroscopy, SEM, TEM, TGA/DTG, VSM, and BET analysis. The effects of various parameters such as the amount of crosslinker, initiator, Fe₃O₄, and reaction time as well as monomer ratio on the oil absorption of the organogel were studied. The synthesized organogel can absorb about 35.5, 22.1, 29.86, 14.58, 17.6, 15.3, and 13.7 g·g⁻¹ of CHCl₃, toluene, CH₂Cl₂, hexane, crude oil, gasoline, and diesel oil, under the optimized polymerization conditions, respectively. The nanocomposite organogels can be easily separated by a magnetic field after absorption of organic solvents.

Keywords Nanomagnetic organogel; Dodecyl methacrylate; Styrene; Crosslinking; MWCNT

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INTRODUCTION

Water pollution caused by organic pollutants and petrochemical products has been considered as an extensive environmental problem which needs special attention. Even very low concentrations of organic solvents and oils can be toxic to microorganisms responsible for biodegradation of sewage processes. Various methods including physical (*e.g.* sorbents, skimmers), chemical (*e.g.* burning, solidifiers), and biological methods have been used to clean up oil spills.^[1,2] Absorption by organogels can be utilized for efficient removal of the contaminants, although the method may impose a limitation on the cost of the starting materials. Low cost porous materials such as clays, zeolites, silica, activated carbon, and natural fibers can be used, but the materials have some disadvantages such as relatively low absorption capacities and low hydrophobicity.^[3–8] In addition, they are not reusable and have no response to environmental stimuli. By contrast, smart organogels can be designed so that they respond to external stimuli. For instance, magnetic organogels can be easily separated by a magnetic field after absorption of organic solvents. Therefore, several modifications of the materials have been developed.^[9–16] A method that provides an alternative route for synthesis of organogels is the use of lipophilic monomers in the polymerization

process.^[17–19] In recent years, novel organogels have been prepared based on micro/nano-materials, polymerizable ionic liquids, polymeric hybrid materials, and waste recycled rubber.^[20–37] It is very difficult to prepare an absorbent that have all the properties required for ideal sorbent including hydrophobicity, high absorption capacity, high rate of uptake, good selectivity, durability in aqueous media, reusability, and biodegradability. Therefore, academic and industrial research should continue to develop the new oil absorbent materials. In this regard, magnetic materials have been paid with much attention because of their special response to external magnetic field in medium.^[38–42]

In this work, a nanomagnetic organogel as a new absorbent material was synthesized using DDMA and styrene monomers *via* emulsion polymerization. DDMA is a very useful feedstock for syntheses of organogels, because it readily undergoes polymerization reactions with a wide variety of organic and inorganic compounds. The design of advanced materials using aromatic monomers with enhanced π stacking properties also benefits the adsorption of aromatic hydrocarbons. The oil absorbency, thermal stability, and hydrophobicity of the organogel as well as its reusability were investigated. The application of these nanomagnetic organogels for oil-water separation is demonstrated, so are their recovery from pollutants and recyclability. The prepared oil-absorbent was readily removed by a magnetic field after absorption of organic solvents. The results show that the nanomagnetic organogel has a good oil absorbency in comparis-

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on with other absorbents.

EXPERIMENTAL

Material

Dodecyl methacrylate (DDMA) and multi-walled carbon nanotube (MWCNT) were purchased from Sigma-Aldrich. Styrene (St) as monomer, divinylbenzene (DVB) as cross-linking agent, and azobisisobutyronitrile (AIBN) as initiator, were obtained from Merck Millipore. Iron oxide nanoparticles (Fe_3O_4) (98%, 20–30 nm) were purchased from US Research Nanomaterials. Sodium dodecylbenzene sulfonate (SDBS) was obtained from Daejung chemicals, Korea.

Instrumental Characterization

Fourier transform infrared (FTIR) spectra were recorded in the form of KBr pellets using a Tensor 27, FTIR spectrophotometer (Germany). Thermogravimetric analysis (TGA) was performed using a Perkin Elmer, Pyris Diamond analyzer (USA) under N_2 atmosphere at a scan rate of 20 °C/min. The surface morphology of the organogel was studied using a scanning electron microscope (SEM) EM 3200, KYKY (USA). Transmission electron microscopy (TEM) was taken on a Zeiss EM10C TEM at an acceleration voltage of 80 kV. The magnetization of the nanomagnetic organogel was measured at room temperature using a vibrating sample magnetometer (VSM, Model MDKB, Iran).

Organogel Synthesis

The oil-absorbents were prepared by *in situ* polymerization-crosslinking method. Briefly, dodecylmethacrylate (3 mL) and styrene (1 mL) were mixed with 0.1 g of AIBN initiator and various weight percentages of crosslinker (DVB, ranging from 0.6 wt% to 8.2 wt%). The reaction mixtures were introduced into a 20 mL flask and allowed to polymerize at 75 °C for 18 h.

Preparing Nano-magnetic Organogel

A typical experiment for the synthesis of nano-magnetic organogel is as follows: 250 mg of Fe_3O_4 and various weight percentages of MWCNT ranging from 0.11 wt% to 0.9 wt% were dispersed into 50 mL of deionized water with sonication for 5 min. Then, 500 mg of sodium dodecylbenzenesulfonate (SDBS) and 1 mL of DDMA were sequentially added into the solution under continuous sonication for another 10 min. The final mixture was then transferred into a three-necked flask which was equipped with a magnetic stirrer, a condenser, and a nitrogen gas inlet. The mixture was stirred at 30 °C for 20 min while being purged with nitrogen. After that, the temperature was raised up to 75 °C, and then a mixed solution of DDMA (3 mL), St (1 mL), DVB (0.05 mL), and AIBN (100 mg) was dropwise added to the solution for 10 min, and the reaction was allowed to continue for 7 h at 75 °C to form a stable emulsion system.

Absorbency Measurement

Swelling properties of the resultant organogels were studied by weighing the resultant gels swollen in different organic solvents as a function of time. The dried gel was cut into small pieces. The sliced gel was placed in 10 mL of organic solvents at room temperature. After a certain interval of time,

the swollen gel was taken out and weighed. The swelling degree (Q) was calculated by the following formula:

$$Q\left(\frac{\text{g}}{\text{g}}\right)=\frac{w_s-w_d}{w_d} \quad (1)$$

where w_d and w_s are weights of the dried gel and the swollen gel after immersion in solvent for a certain time, respectively.

Absorption Kinetics of Organogels

To study the absorption kinetics, organic solvent absorbency was investigated at different time and the experimental data were further analyzed using kinetic models.

For the first-order kinetic model, the swelling rate is expressed as Eq. (2):

$$\frac{dQ}{dt}=k_1(Q_{\max}-Q) \quad (2)$$

where $Q_{\max}$ is the maximum organic solvent absorbency of the gel, Q is the absorbency at time t , and k is the swelling kinetic constant. If plotting the integration form of Eq. (2), $\ln[Q_{\max}/(Q_{\max}-Q)]$ versus t gives a straight line, the swelling process follows the first-order kinetic model.

$$\ln\left(\frac{Q_{\max}}{Q_{\max}-Q}\right)=k_1t \quad (3)$$

For the second-order kinetic model, the rate of the swelling process is shown as the following equation:

$$\frac{dQ}{dt}=k_2(Q_{\max}-Q)^2 \quad (4)$$

This equation is displayed after integrating as follows:

$$\frac{t}{Q}=\frac{1}{k_2Q_{\max}^2}+\frac{1}{Q_{\max}}t \quad (5)$$

If the plot of t/Q versus t is a straight line, the swelling tends to follow the second-order kinetic model.

RESULTS AND DISCUSSION

Synthesis and Characterization

The nanocomposite organogel was prepared by *in situ* emulsion polymerization-crosslinking method using dodecyl methacrylate (DDMA) and styrene (St) as the monomers, azobisisobutyronitrile (AIBN) as an initiator, and divinylbenzene (DVB) as a crosslinking agent. A proposed mechanism is shown in Fig. 1. AIBN radically initiated the polymerization of DDMA and St. A three-dimensional network could be formed in the presence of DVB as a crosslinking agent. Aliphatic solvents can interact with the non-polar long-chain of DDMA. The presence and interaction of Fe_3O_4 nanoparticles and MWCNT are indicated schematically in Fig. 2. Fe_3O_4 nanoparticles can interact with polar functional group of DDMA. MWCNTs can interact with the non-polar long-chain of DDMA. A non-covalent π - π interaction with the phenyl group of styrene can be established in the presence of aromatic solvents such as benzene and toluene.

FTIR spectra of the samples are shown in Figs. 3(a) and 3(b). In the spectrum of organogel, the absorption bands at 2922 and 1726 cm^{-1} are attributed to the C–H stretching mode of alkyl chain and C=O stretching mode of the ester

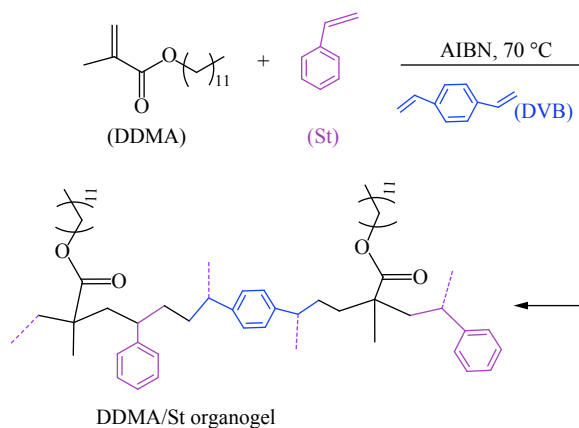


Fig. 1 Schematic representation of preparation of DDMA/St organogel

groups, respectively. In the FTIR spectra of the nanomagnetic organogel, the intensity of absorption band at 1726 cm^{-1} decreased due to the interaction of carbonyl group with iron oxide nanoparticles, as indicated in Fig. 3(b).

TGA/DTG thermograms of the samples are demonstrated in Figs. 3(c) and 3(d). The nanomagnetic organogel showed higher thermal stability than the organogel did. This can be attributed to the presence of iron oxide nanoparticles and

MWCNT. The weight reduction at $600\text{ }^{\circ}\text{C}$ was about 93% for the nanocomposite organogel. DTG indicates that the rate of organogel degradation at $360\text{ }^{\circ}\text{C}$ was higher than that of nanocomposite organogel degradation.

The SEM images of freeze-dried samples were taken to study the surface morphology of the organogels. As indicated in Figs. 4(a) and 4(b), both the organogel and the nanomagnetic organogel had porous structures, but the types of holes and porosity were different.

TEM analysis was used to study the accumulation and dispersion of nanoparticles in the organogel matrix. As shown in Figs. 4(c) and 4(d), the nanomagnetic particles were well dispersed in the organogel matrix. The presence of the nanotubes is also clearly visible in these images.

VSM analysis shows a single domain of magnetic nanoparticles formed within the organogel networks and the superparamagnetic behavior of the nanomagnetic organogel (Fig. 5). A hysteresis loop with a saturation magnetization (M_s) of 3 emu/g was observed. This M_s value is much lower than that of the original magnetic nanoparticles, but is expected due to the presence of only 3.5% of magnetic nanoparticles loaded in the network.

Absorption Studies

The absorption of organic solvents was studied as a function

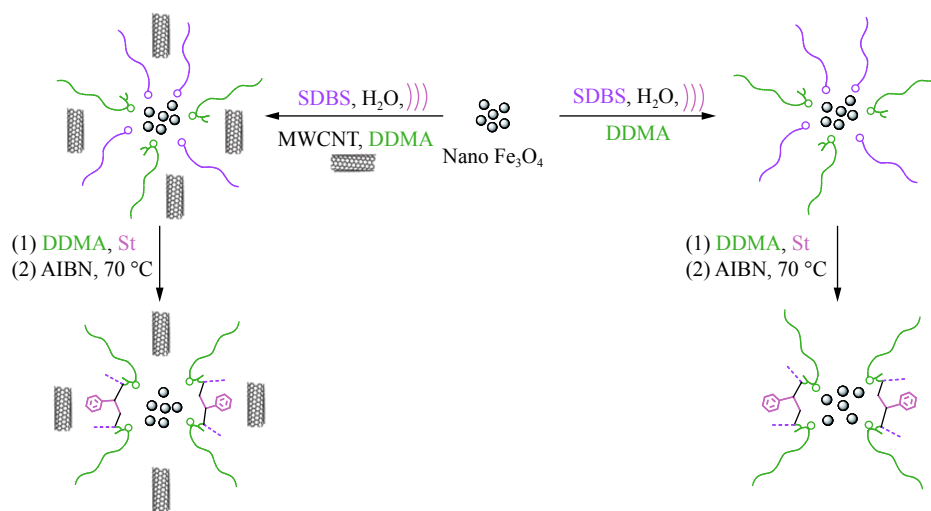


Fig. 2 Schematic representation of preparation of nanomagnetic organogels

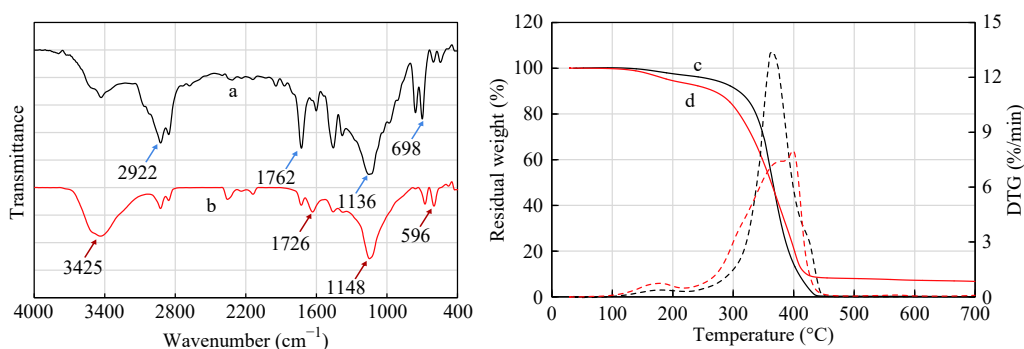


Fig. 3 FTIR spectra of (a) the DDMA/St organogel and (b) nanomagnetic organogel; TGA/DTG thermograms of (c) the organogel and (d) nanomagnetic organogel

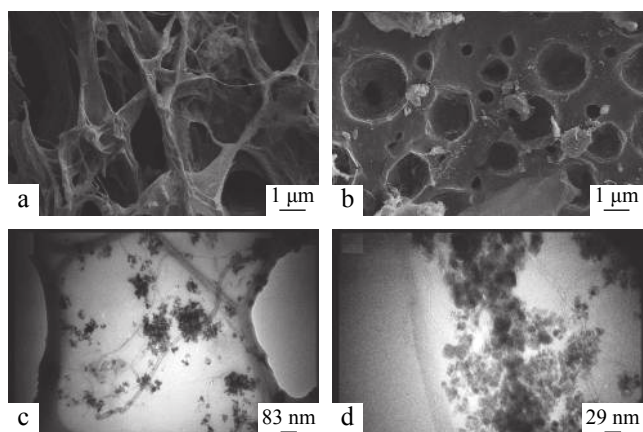


Fig. 4 SEM images of (a) the organogel and (b) nanomagnetic organogel, and (c, d) TEM images of the nanomagnetic organogel with different magnifications

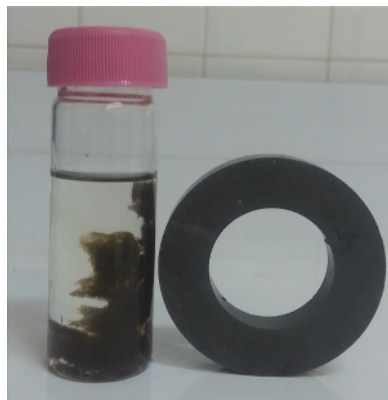
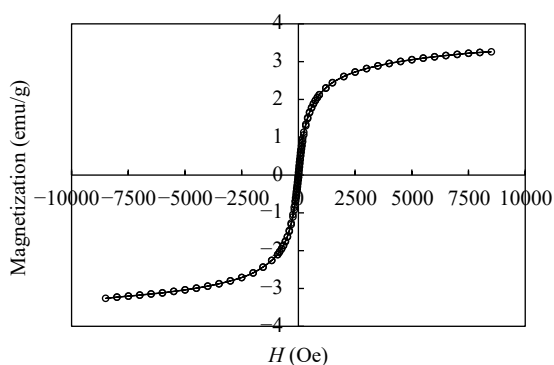


Fig. 5 VSM analysis of the nanomagnetic organogel

of time. The swelling ability of nanomagnetic organogel can be attributed to the lipophilicity of the dodecyl groups of DDMA and MWCNT, as well as the interaction of solvents with DDMA ester group and nanomagnetic particles. As can be seen from Fig. 6, the maximum absorption was observed for chloroform. The chemical selectivity depended on the interaction between solvent molecules and the polymer chains, and also the diffusion of solvent molecules into the organogel.

The nanomagnetic organogels were prepared by inclusion of nanomagnetic particles in the polymer matrix. The amount of nanomagnetic particles in polymer matrix was varied and

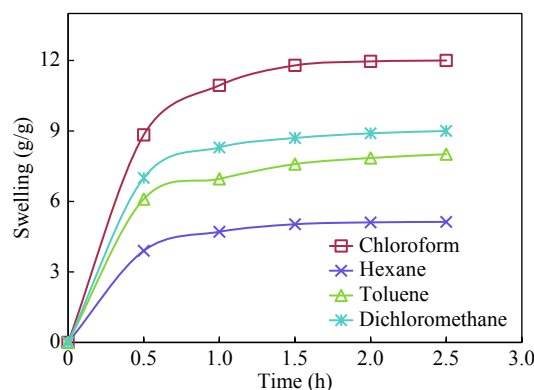


Fig. 6 Absorption of organic solvents by the nanomagnetic organogel in the presence of DVB

its effect on absorption of solvent was studied (Fig. 7a). There was no substantial change in absorption of organic solvent when the amount of magnetic nanoparticles was varied from 1.5% to 3.5%. It can be attributed to the interaction between magnetic nanoparticles and polar groups of polymer matrix. However, a further increase in the amount of nanoparticles led to a decrease in solvent adsorption, which was due to the hydrophilicity of the magnetic nanoparticles.

The presence of a crosslinker is essential for successful synthesis of polymeric network, and incorporation of an appropriate crosslinker affects the swelling properties of the final product. Nanomagnetic particles in addition to DVB can also act as crosslinker *via* an ion-dipole interaction. It was found that the absorption of solvent increased substantially in the absence of DVB (Fig. 7b). The higher adsorption of toluene and benzene solvents can be attributed to the better diffusion of the solvent molecules into the organogel matrix. Comparison between chloroform absorption in the presence or absence of DVB illustrates this point, as shown in Fig. 7(c). The effect of CNT addition was further examined (Fig. 7d). CNTs have a unique structure, large surface area, and oleophilic properties. In addition, the mechanical properties of nanocomposites can be considerably improved by the use of CNT. The absorption of organic solvents decreased with the increasing amount of MWCNT (Fig. 7d). This can be attributed to the formation of an inappropriate or ineffective network in the presence of an excess amount of MWCNT. MWCNT may also play the role of crosslinker. The absorption of crude oil, gasoline, and diesel oil from water surface was investigated using the nanomagnetic organogels (Fig. 8a). As shown in Fig. 8(b), the nanomagnetic organogel can absorb about 17, 15, and 13 g·g⁻¹ of gasoline, crude oil, and diesel oil from water surface, respectively.

Absorption kinetics of organic solvents by the nanomagnetic organogel were studied. The plot of t/Q versus t is shown in Fig. 8(c). The best fitting was found for the second order model. So, the absorption followed the second order kinetic mechanism. The calculated kinetic parameters are presented in Table 1.

The reusability of the nanomagnetic organogel was investigated. The nanomagnetic organogels were dried at ambient temperature after solvent adsorption, and then applied for

re-absorption of the organic solvents. The results are shown in Fig. 8(d). A significant decrease was observed for absorption of organic solvent in the second stage which is probably due to the change in the organogel structure. However,

these nanomagnetic organogels still can be used for several times.

Table 2 illustrates the properties of some oil adsorbents in comparison with the nanomagnetic organogel. The oil ad-

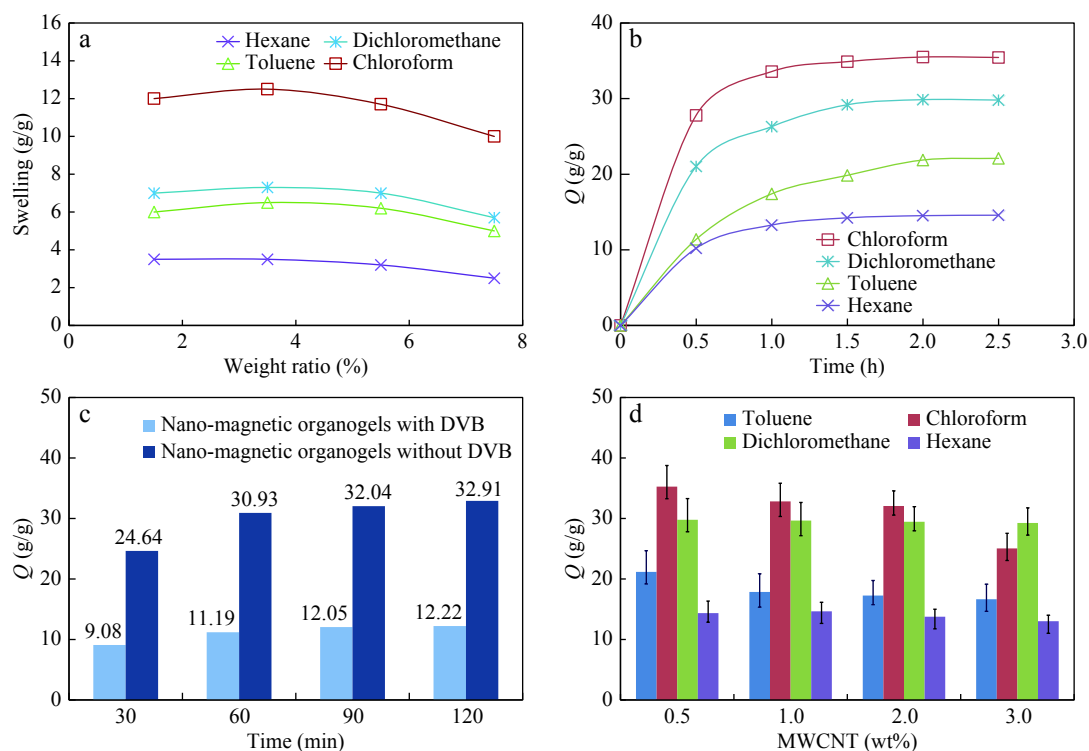


Fig. 7 (a) Variation of the nanomagnetic particles in polymer matrix; (b) Absorption of organic solvents by the nanomagnetic organogel in the absence of DVB; (c) Comparison of chloroform absorption by the nanomagnetic organogel in the presence or absence of DVB; (d) Absorption of organic solvents in the presence of MWCNT

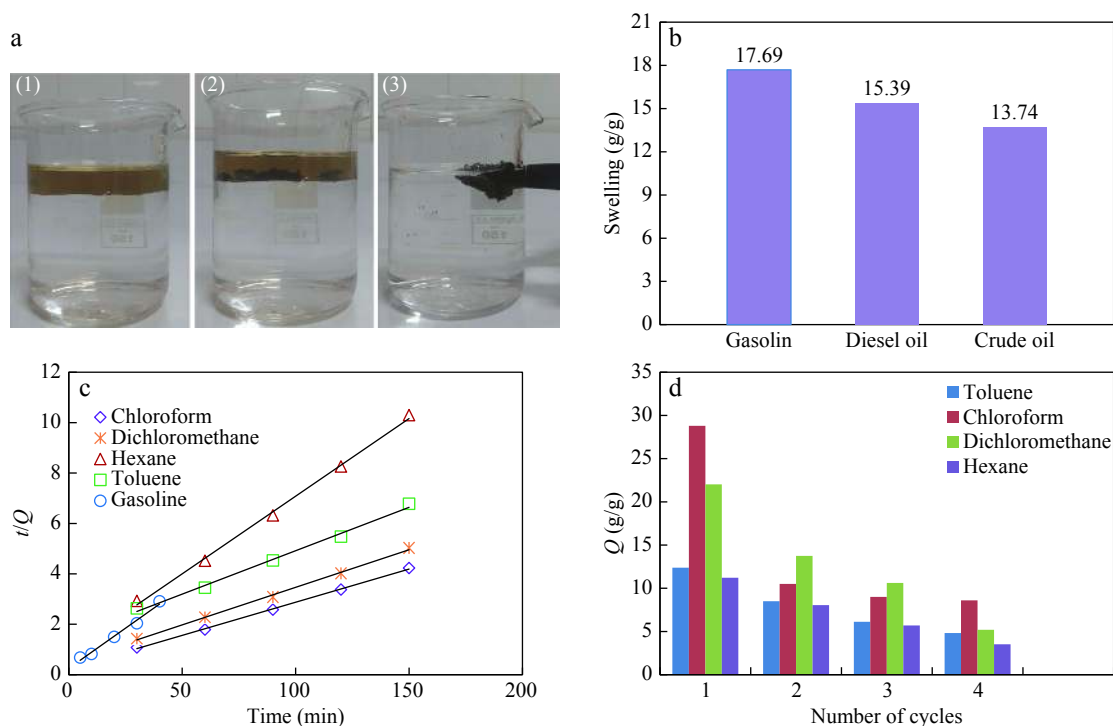


Fig. 8 (a, b) Absorption of crude oil, gasoline, and diesel oil from water surface by the nano-magnetic organogel; (c) The plot of t/Q versus time of the second order model; (d) The reusability of the nano-magnetic organogels for solvent absorption

Table 1 Kinetic parameters of absorption of organic solvent

Organic solvents and oils	$Q_{\max}$	k_2 (g·g ⁻¹ ·min ⁻¹)	R^2
Chloroform	35.5	0.0028	0.999
Toluene	22.1	0.0009	0.984
Dichloromethane	29.86	0.0018	0.998
Hexane	14.58	0.0041	0.998
Oil	13.39	0.0012	0.997
Gasoline	14.69	0.016	0.987
Euro diesel	11.74	0.00025	0.948

sorbents can have different forms and shapes such as granules, fiber, sponges, gel, etc. It is important to develop a low cost oil adsorbent that has a high rate of sorption, with high capacity. In addition, the processes of its recovery should be economical and practical. However, no adsorbent has all the above characteristics. The synthesized nanomagnetic organogel can be easily separated by magnetic field and recovered at room temperature.

Table 2 Comparison of some properties of various oil adsorbents

Type of absorbent and structure	Type of oil/solvent	Method of testing and capacity range (g·g ⁻¹)	Reusability	Magnetic separation/ Recovery method	Ref.
Graphene aerogel/nano-microporous network	Diesel oil, toluene, cyclohexane, acetone	Immersion into organic solvents, 40–250	After 10 cycles, 91%	No/Distillation	[43]
Chitosan-cellulose aerogel/macroporous network	Diesel, gasoline, carbon tetrachloride, acetone	Immersion, and oil/water mixture 15–28	After 50 cycles, 75%	No/Squeezing	[10]
Acetylation of banana fiber/fiber	Machine oil	Mixed with oil-water suspension, 7–18	After 3 cycles, 40%	No/Squeezing	[44]
Porous Al ₂ O ₃ -acrylic resin/porous resin	Edible oil, engine oil, toluene, carbon tetrachloride	Immersion into organic solvents, 6–30	After 5 cycles, 96%	No/Extraction, vacuum drying	[32]
Magnetic silica nanocomposite/nano-composite particles	Edible oil, diesel oil, toluene, cyclohexane, hexane	Immersion into water-oil mixture, 4–7	After 20 cycles, 90%	Yes/Washing, air-drying	[45]
Multi-walled carbon nanotube/nano tube	Gasoline, diesel oil, kerosene	Mixing the adsorbent with oil, 4–19	After 10 cycles, about 50%	No/Squeezing and heat treatment	[46]
Magnetic-silanized ethyl cellulose sponges/porous sponges	Diesel oil, hexane, gasoline, petroleum ether, soybean oil	Immersion into the organic liquids, 30–50	After 10 cycles, about 90%	Yes/Heating at the boiling point and distillation	[47]
Chitosan-PAN-magnetic ZSM-5 zeolite coated sponges/nanofibrous sponges	Pump oil, lubrication oil, motor oil	Tested on water surface, 45–95	After 5 cycles, about 95%	Yes/Squeezing, and drying under vacuum	[48]
Nano-magnetic organogels based on DDMA/porous organogel	Gasoline, crude oil, diesel oil, toluene, chloroform	Immersion and tested on water surface, 13–35	After 4 cycles, 30%	Yes/Drying at RT	This work

CONCLUSIONS

A novel nanomagnetic organogel was synthesized by *in situ* emulsion polymerization-crosslinking using dodecyl methacrylate (DDMA) and styrene (St). The structure of nanomagnetic organogel was modified by inclusion of MWCNT into the matrix. Absorption experiments of organic solvents showed that the nanomagnetic organogels absorb about 35.5, 22.1, 29.86, and 14.58 g of CHCl₃, toluene, CH₂Cl₂, and hexane, respectively. The results also indicated that the nanomagnetic organogel could absorb about 17.6, 15.3, and 13.7 g·g⁻¹ of gasoline, crude oil, and diesel oil from water surface, respectively. The absorption followed the second order kinetic mechanism. The reusability of absorption experiment demonstrated that the nanomagnetic organogels could be used for several times.

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