Rapid Communication

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# Recent advances in photothermal materials for solar-driven crude oil adsorption

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**Abstract:** In recent years, the adsorption method is usually adopted in the actual treatment of crude oil spills. However, the high viscosity of crude oils prevents them from diffusing into the internal pores of the adsorbent, resulting in ineffective oil capture. Photothermal materials can reduce the viscosity of crude oil by *in situ* heating through the photothermal conversion effect, making it easier for crude oil to occupy the internal pores of the adsorbent. At present, the review of the application of photothermal materials in the field of crude oil adsorption is still blank. This review focuses on the application of novel photothermal conversion materials in the field of crude oil adsorption and their performance comparison. Among the photothermal conversion materials used in the field of crude oil adsorption, some are commercial sponges with high porosity and photothermal coating, while others are self-assembled three-dimensional porous structures of materials with inherent photothermal properties. This review mainly introduces the types and research progress of materials with good photothermal effect at home and abroad in recent years and summarizes some new research ideas and materials that can be applied to photothermal conversion.

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**Keywords:** sunlight utilization, photothermal conversion mechanism, photothermal conversion materials, crude oil adsorption

## 1 Introduction

Oil is an indispensable resource for human production. In the process of oil exploration, recovery, and transportation, oil leakage will cause significant harm to the ecosystem [1,2]. Various efforts, such as gravity separation, oil boom isolation, solidification, dispersion, bioremediation, and combustion [3–7], have dealt with crude oil spills. However, these methods often lead to massive energy consumption, resources, and time [8]. They are also inefficient for crude oils with high viscosity and low mobility, and the chemicals used or produced in the process sometimes cause secondary pollution [9]. Adsorption is a simple, efficient, and effective method for dealing with oil spills without secondary pollution [10]. The porous photothermal material can be used for crude oil spill repair due to its adsorption, photothermal conversion, and robust reproducibility. It does not negatively impact the environment when used for *in situ* heating to harvest crude oil from the ocean surface or oil/water mixtures.

In recent years, more and more studies have been conducted on light-absorbing materials to apply the solar thermal conversion to crude oil adsorption. Metal-based nanoparticles and inorganic semiconductor materials are rarely studied in crude oil adsorption because they do not have a pore structure and only have light absorption properties at some wavelengths. They are usually used in photothermal therapy and solar water evaporation. Two mainstream 3D network photothermal conversion structures are currently studied in crude oil adsorption: the first is the interconnected commercial sponge, which has been widely studied due to its low cost, high porosity, and robust mechanical properties. However, it has no self-heating performance, and its photothermal conversion performance can only be improved by coating graphene [11], dopamine [12,13], and other photothermal coatings. The practical application is achieved by coating the hydrophobic and thermally conductive layers [14]. The second is to use graphene and its derivatives [15–17], carbon nanotubes (CNTs) [18,19], MXene materials [20], *etc*., to self-assemble into framework materials with self-healing properties. Then, the photothermal elements (such as metal coordination compounds [21–23] and inorganic semiconductors [24,25]) are loaded to improve their photothermal conversion performance further. Therefore, this article mainly reviews recent advances in photothermal materials for solar-driven crude oil adsorption.

# 2 Photothermal conversion mechanism

Photothermal materials capable of harnessing solar energy have attracted great interest in the past few decades. An ideal photothermal conversion material should have broad-spectrum solar absorption and high photothermal conversion efficiency. Under sunlight irradiation, the material absorbs photons in the sunlight, causing it to be photoexcited. Driven by photoexcitation, hot electrons are generated, resulting in the distribution of thermal charge flow, which ultimately converts solar energy into thermal energy. According to the type of electrons and the bandgap structure, the photothermal conversion mechanism can be divided into the following three categories: (1) Localized Surface Plasmon Resonance (LSPR) effect, (2) electron–hole generation and nonradiative relaxation, and (3) conjugation or hyperconjugation effect.

## 2.1 LSPR effect

Common metal-based nanoparticles have the LSPR effect [26]. When the diameter of metal nanoparticles is smaller than the excitation wavelength, an electric field of a specific frequency will cause the coherent oscillation of metal-free electrons when they pass through the nanoparticles. Optical radiation induces electrons to oscillate from an occupied state to an unoccupied state, generating hot electrons that are converted into thermal energy. The lattice then transfers heat to the surrounding medium through phonon–phonon coupling, thereby raising the surrounding temperature. Gold, silver, and platinum nanoparticles are the most common plasmonic metals for photothermal conversion applications. It has been reported that when the plasmon resonance wavelength of gold is equal to the wavelength of laser illumination, its temperature can reach above 70°C [27].

## 2.2 Electron–hole generation and nonradiative relaxation

Electron–hole generation and nonradiative relaxation of semiconductors generally exist in inorganic semiconductor materials. When the incident light is greater than the bandgap energy of the semiconductor, strong absorption occurs at wavelengths where the bandgap energy matches, generating electron–hole pairs [28] equivalent to the bandgap energy. When the excited electrons return to the lowest energy level, there are two release paths. One part is released in the form of photons by radiative relaxation [29]; the other part is released in the form of phonons by nonradiative relaxation, and the phonons interact with the lattice to generate heat. This mechanism applies to various narrow-bandgap semiconductors, such as CuS [30] and $MoS_2$ [31]. For wide-bandgap semiconductors, most of the absorbed energy is re-emitted in the form of photons, which are prone to electron–hole pair recombination.

## 2.3 Conjugation or hyperconjugation

In a conjugated system, the overlapping of adjacent π electrons or the interaction of π bonds with p orbital electrons redistributes the electron density, resulting in conjugation effects. While most chemical bonds, such as C–C, C–H, O–H, and C–O, exist as σ bonds, the interaction between the electrons of the σ bond and the adjacent empty orbitals or partially filled p orbitals also produces a conjugation effect called the hyperconjugation effect. The conjugation effect and hyperconjugation effect make electrons have strong light absorption in the near-infrared region and accelerate the migration between electrons. These electrons can be excited from π to π* orbitals with lower energy inputs, releasing heat on their return to the ground state. These two effects enable the photothermal conversion of many carbon-based materials with conjugated structures, such as graphene and CNTs. In addition, some organic polymers with conjugated structures, such as polydopamine (PDA) and polypyrrole (PPy), also have photothermal properties. The excited state electrons relax from the lowest occupied molecular orbital to the highest occupied molecular orbital through electron–phonon coupling, thereby generating thermal energy.

# 3 Crude oil adsorption

## 3.1 Liquid adsorption mechanism

For the liquid adsorption process of three-dimensional porous materials, the adsorption efficiency is mainly determined by the porosity, surface wettability, absorbent pore size, average tortuosity factor, and liquid viscosity. The adsorption efficiency increases with increasing adsorbent porosity, pore size, surface tension, decreasing liquid viscosity, and decreasing tortuosity of the adsorbent [32]. Based on the liquid adsorption mechanism, we can improve the oil absorption efficiency from the following three aspects to achieve the purpose of adsorbing crude oil: (1) hydrophobic/oleophilic modification; (2) enhancement of the inner capillarity action of oil adsorption materials (tortuosity reduction); and (3) combination of heating methods with oil adsorption materials [33].

## 3.2 Application of photothermal conversion to crude oil adsorption

Generally speaking, the composition of crude oil is very complex. Due to their molecular structure [34] and the influence of other heteroatoms or groups [35], they have such high viscosity that they are difficult to be adsorbed by ordinary porous adsorbents. The viscosity of crude oil is a key parameter affecting its recovery, production and transportation, and remediation [36]. The viscosity of crude oil is largely dependent on two external factors: temperature and pressure. Elevated temperature causes crude oil to become less viscous and increase its fluidity. Photothermal conversion materials can use their own characteristics to convert absorbed sunlight into heat energy to heat crude oil *in situ* to reduce the viscosity of crude oil, so that it is easier to be adsorbed. Therefore, this review mainly focuses on the use of solar energy to assist adsorbents with photothermal effect to achieve high-efficiency photothermal conversion to accelerate the crude oil adsorption.

# 4 Research status of new photothermal conversion materials and their crude oil adsorption

With the development of science, technology, and human society, the demand for clean energy utilization in various fields has increased. Photothermal materials with low price, easy functionalization, easy preparation, and broad spectral absorption have been widely studied. At present, common new photothermal conversion materials mainly include: metal-based nanoparticles [37], inorganic semiconductor materials [38], organic polymers [39–41], Mxene materials [42,43], and carbon-based materials [44,45]. Porous photothermal conversion materials have been widely studied in crude oil adsorption applications.

## 4.1 Metal-based nanoparticles

Metal-based nanoparticle photothermal conversion materials are mainly silver, gold, palladium, and other precious metals. Surface plasmon oscillations of metal electrons lead to enhanced light absorption in electromagnetic fields. The surface plasmon absorption spectrum undergoes a red shift as the particle size increases. Nanoparticles of the right size can strongly absorb visible hooks and near-infrared light. Therefore, the photothermal conversion properties of these metal-based nanoparticles are affected by their shape, size, and environment. At the same time, its light absorption range is limited, and it is generally used in conjunction with other photothermal materials with broad spectral light absorption. Metal-based nanomaterials are usually only supported simply. It is easy to be detached during use, which greatly reduces the light-to-heat conversion performance of the material. Shi *et al.* [46] prepared magnetic lignin-based polyurethane foam by adding polyurethane and $Fe_2O_3$ nanoparticles during lignin foaming. The prepared foam has good photothermal conversion and recycling performance and can be used to remove viscous crude oil in water. Ding *et al.* [47] reported a UV-initiated crosslinking of polysodium methacrylate (pNaMMA), then replaced $Na^+$ ions with $Ag^+$, and then photothermally *in situ* reduced to silver nanoparticles (AgNPs). The prepared pNaMMA/AgNPs fabric has the effect of oil–water separation and sterilization. The materials reported by them all make the matrix material have strong bonding with metal-based nanoparticles and have good recycling performance.

## 4.2 Inorganic semiconductor materials

Black inorganic semiconductor materials have become the research focus of photothermal conversion materials due to their advantages of various types and easy functional modification, usually including black titanium dioxide, $Cu_7S_4$, $Ti_3AlC_2$, *etc*. Generally speaking, inorganic semiconductors are commonly used in fields, such as photothermal therapy [48] and solar water evaporation [49], due to their excellent processability and biocompatibility [50]. Inorganic semiconductors are rarely used in crude oil adsorption and must be combined with other materials. Inorganic semiconductor materials are similar to metal-based nanoparticles. In the field of crude oil adsorption, attention should also be paid to the problem of photothermal unit falling off. Li *et al.* [51] used carbon black nanoparticles to decorate the viscous polymer foam skeleton, which could be heated to more than 80°C under the irradiation of 1 Sun, and the adsorption capacity reached 6 g/g. Sun *et al.* [52] assembled $CuFeSe_2$ nanoparticles synthesized by wet chemistry with graphene aerogels to synthesize graphene aerogel-$CuFeSe_2$ (GA-$CuFeSe_2$). It has an ultrafast adsorption rate and a large adsorption capacity at a specific wavelength (808 nm). Niu *et al.* [53] deposited CuS nanoparticles on melamine sponges (MSs) (Figure 1a). Under sunlight, the sponge can be quickly heated, effectively reducing the viscosity of the surrounding crude oil and enhancing the fluidity. The peristaltic pump can continuously absorb crude oil at 5.30 g/min. Li *et al.* [54] designed a layered CuO@CuS/PDMS nanowire array (NWA) inspired by the Crassula perforata-Structured (Figure 1b). This structure has excellent photothermal conversion performance and thermal conductivity, and the adsorption capacity of crude oil can reach $1.56 \times 10^6$ g/m$^3$ in the adsorption process of 5 min.

## 4.3 Organic polymers

Common organic polymer photothermal conversion materials mainly include dopamine and PPy. The construction of a donor–acceptor structure within the dopamine system resulted in a lower energy gap and increased electron delocalization [55]. The unique wrinkled structure of multilayer PPy is formed spontaneously during the polymerization process. The PPy surface will absorb incident light at different angles after multiple reflections, promoting the broad-spectrum light capture capability of the multilayer PPy nanosheets [56]. Compared with metal-based nanoparticles and inorganic semiconductors, which have photothermal conversion effects only in a certain wavelength range, organic polymers have the ability to capture broad-spectrum light. While sunlight has a very wide wavelength range, organic polymers can better utilize the energy of sunlight and can be better applied in practice. Thence, these advantages make them suitable for crude oil adsorption. While organic polymers need to pay attention to their environmental stability, they are prone to corrosion and photodegradation. Therefore, they usually need to be protected by other functional coatings in practical use. In 2015, Wu *et al.* [57] integrated the sunlight-induced photothermal conversion effect of PPy and the thermoresponsive properties of poly(*n*-isopropyl acrylamide) (PNIPAm) into an MS. The material successfully achieved rapid absorption of heavy oil under sunlight and passive oil release underwater at room temperature. In 2018, Zhang *et al.* [58] synthesized a self-heating hydrophobic/lipophilic sponge by depositing PDA and polydimethylsiloxane using an aqueous deposition process, benefiting from the photothermal conversion effect of PDA coating. The temperature of the sponge is rapidly increased, reducing the viscosity of the crude oil *in situ*. The adsorption capacity of the self-heating sponge can reach $1.29 \pm 0.37 \times 10^6$ g/m$^3$. At the same time, they integrated a self-heating sponge with a peristaltic pump to create a self-heating vacuum cleaner (Figure 2a) that enables continuous cleaning or collection of crude oil from the water surface. In 2021, Li *et al.* [59] constructed a polydimethylsiloxane (PDMS)/polyaniline (PANI)-modified MS by simple polymerization and dip coating (Figure 2b). The unique photothermal coating makes the surface equilibrium temperature rise rapidly to 81.80°C within 2 min,

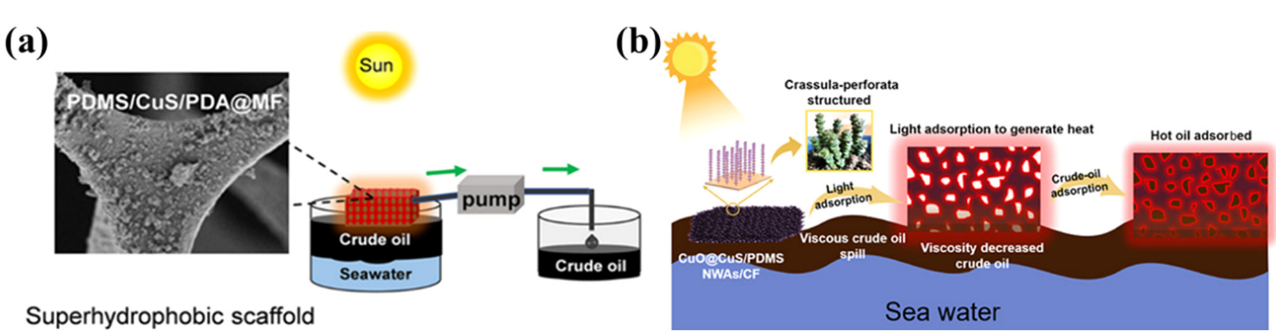


**Figure 1:** (a) Solar-assisted *in situ* crude oil recovery [53]. (b) CuO@CuS/PDMS NWA growth and crude oil adsorption diagram [54].

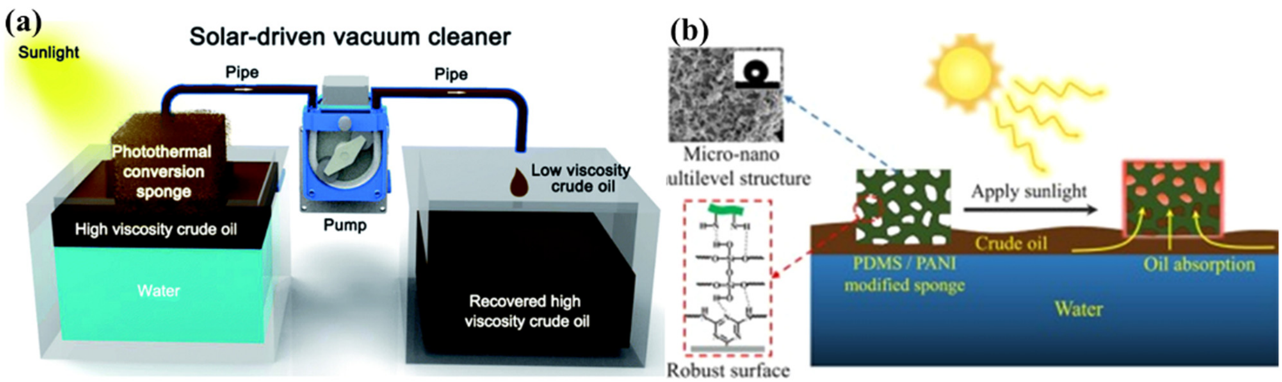


**Figure 2:** (a) Schematic illustration of the solar-driven vacuum cleaner [58]. (b) Schematic diagram of PDMS/PANI photothermal conversion crude oil adsorption [59].

thus showing an excellent adsorption capacity of $1.17 \times 10^6$ g/m$^3$. In 2022, Zeng *et al.* [60] prepared a superhydrophobic cotton fabric with a photothermal conversion effect by the PPy deposition method. The surface of the modified cotton fabric was heated to 68.20°C through the photothermal conversion effect to reduce the viscosity of crude oil and selectively absorb crude oil in water. The efficiency increased from 83.20 to 91.80%.

## 4.4 Inorganic compound MXene materials

The research on inorganic compounds MXene is still in its infancy, and the mechanism is not fully understood. MXene and other materials are necessary for their photothermal behavior due to their excellent solar light absorption properties. At the same time, sunlight can pass through the lattice structure of MXene and can be reflected multiple times between layers, so MXene has efficient light absorption in a wide range of the solar spectrum. MXene is characterized by high mechanical properties, high electrical conductivity, and shape diversity in structure and composition. They are also hydrophobic, so they are also used in crude oil adsorption. Inspired by wood, Cai *et al.* [61] synthesized MXene aerogels with excellent photothermal conversion ability with functionalized cellulose nanocrystals by green mechanochemistry. The desired microstructures can be controllably diversified for structurally adaptive functions through nucleation-driven fine-tuning. MXene aerogels exhibit durable superhydrophobicity, mechanical superelasticity, efficient light oil absorption, and excellent light-to-heat conversion. Ma *et al.* [62] modified lignin-based polyurethane foam with MXene nanosheets (Figure 3a), the maximum equilibrium temperature reached 83.50°C, and the adsorption capacity reached 7.60 ± 0.20 g/g under 1 Sun. At the same time, it is easy to degrade in an alkaline solution, leaving only $TiO_2$ particles that are harmless to the environment, which makes it safer and more environmentally friendly to recycle. Gong *et al.* [63] used a simple strategy to prepare $Ti_3C_2T_X$ Mxene-wrapped commercial sponges

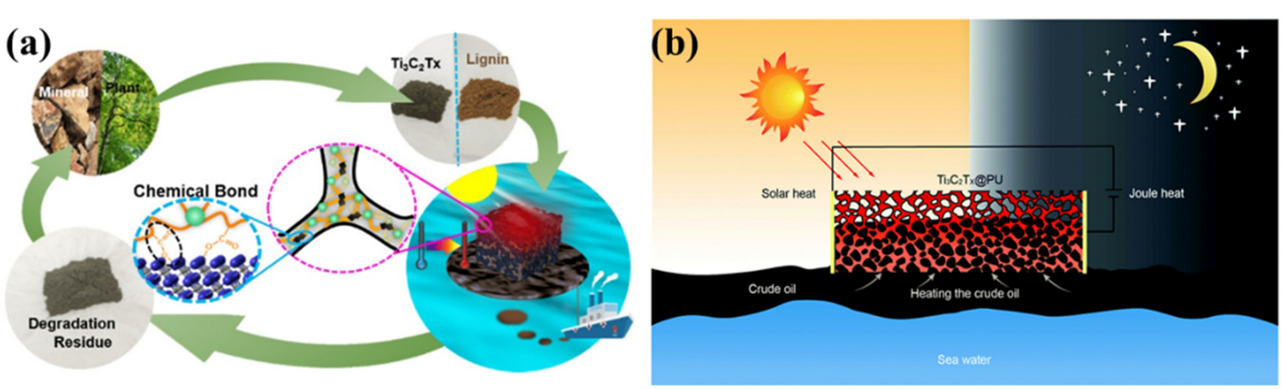


**Figure 3:** (a) Schematic diagram of LPUF photothermal conversion crude oil adsorption and recovery [62]. (b) Schematic illustration of all-weather $Ti_3C_2T_X$@PU used for viscous oil cleanup [63].

($Ti_3C_2TX$@PU) (Figure 3b). Under the irradiation of 1 Sun, the temperature was raised to 75°C within 2 min, and the adsorption capacity reached 43 times its own. The sponge has a good electrothermal conversion effect and can be used for all-weather crude oil adsorption.

## 4.5 Carbon-based materials

Compared with photothermal conversion materials, such as metal-based nanoparticles, organic polymers, MXene materials, or inorganic semiconductors, carbon-based 3D materials have the advantages of broad-spectrum light absorption, excellent photothermal conversion performance, low cost, and tunable pore structure. In addition, the surface of carbon-based materials is easy to expand and modify and can be integrated with various substrates. Carbon-based nanomaterials have been selected as excellent photothermal conversion materials due to their properties and are widely used in crude oil adsorption. Wu *et al.* [64] reported a three-dimensional porous material with a radially arranged porous structure prepared from carbonized natural wood coated with PDMS as the hydrophobic layer (Figure 4a). Low adsorption resistance and high capillary effect make it easier for crude oil to enter the pores of balsa wood for fast cleaning of crude oil. It can raise the surface temperature to 75°C in 1 min under 1.5 Sun. It also compresses and releases at a constant 50% strain for extended periods without structural damage. Luo *et al.* [65] reported a reduced graphene oxide (RGO)-based microsphere aerogel with many radially oriented microchannels (Figure 4b), which was synthesized by growing CNTs within the microchannels and reducing graphene oxide components at high temperature. Due to the efficient photothermal conversion effect and the rough lipophilic surface of the large surface area microchannels, this aerogel promotes the absorption of sunlight, thereby enhancing the adsorption of crude oil. Under 1 Sun, the aerogel surface temperature rapidly rose to 83°C within 1 min. In addition, the aerogel with a large number of radially oriented microchannels has an extraordinary adsorption capacity for heavy crude oil, with an adsorption capacity as high as 267 g/g in 10 min.

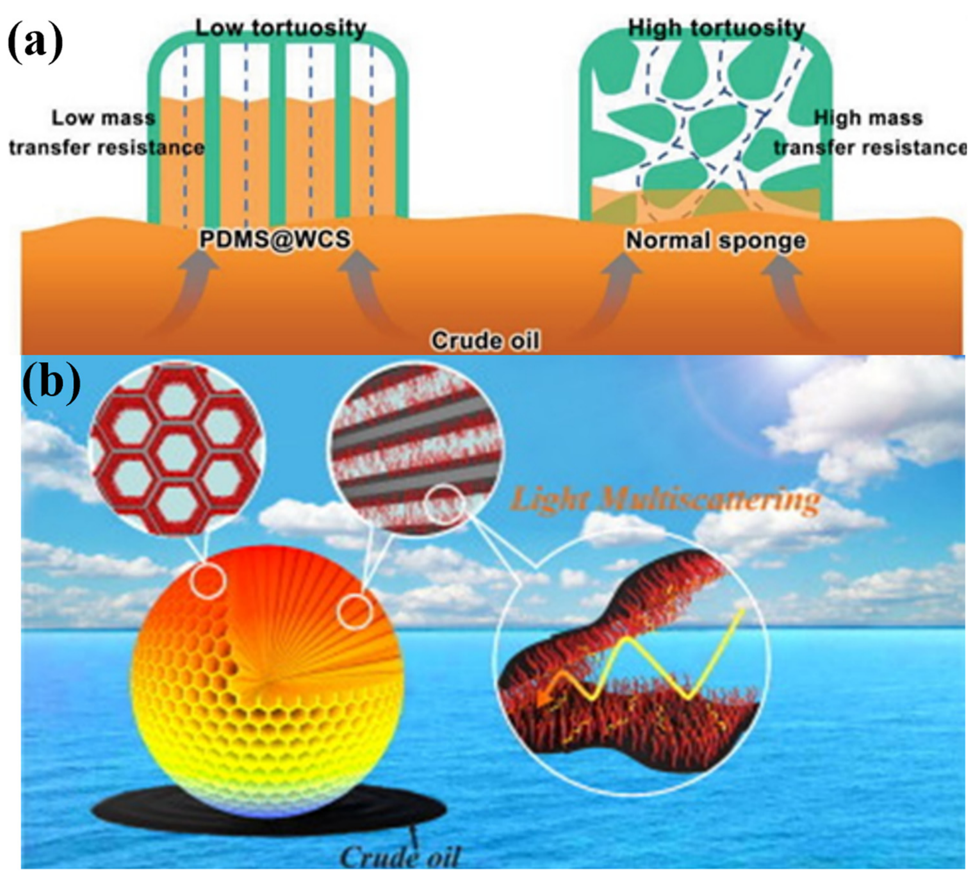


**Figure 4:** (a) Schematic comparison between PDMS@WCS with aligned channels and commercial MS with random pores [64]. (b) Schematic diagram of CNTs/RGO microspherical aerogels photothermal conversion crude oil adsorption CNTs/RGO microspherical aerogels [65].

**Table 1:** Performance comparison of photothermal materials toward removal of oil spills

| Types of photothermal materials | Photothermal conversion materials | Elevated temperature $T$ (°C) | Adsorption capacity | Sorption time $t$ (min) | Desorption method | Recycling time | Intensity of sunlight (kW/m$^2$) | Ref. |
|---|---|---|---|---|---|---|---|---|
| Metal-based nanoparticles | rGO/AgNPs-MS | — | 68 g/g | 5 | Manual squeezing | — | 1 | [66] |
| Inorganic semiconductor materials | GA-$CuFeSe_2$ | 139 | 18.63 g/g | — | — | — | 2.7 | [52] |
| | $MoS_2$-RS | — | 15 g/g | — | — | — | 1 | [67] |
| | CuO@CuS/PDMS | 60 | $1.56 \times 10^6$ g/m$^3$ | 5 | — | 8 | 0.6 | [54] |
| Organic polymers and their composites | PDMS/PDA5 | 80 | $1.29 \times 10^6$ g/m$^3$ | 3 | Mechanical compression | 9 | 1.5 | [58] |
| | PPB@PU | 91 | 45 g/g | — | — | — | 1 | [10] |
| | LPUF30-1 | 90.30 | 6.34 g/g | — | Manual squeezing | 5 | 1 | [68] |
| | PDMS/PANI@MS | 81.80 | $1.17 \times 10^6$ g/m$^3$ | 2 | Manual squeezing | 6 | 1 | [59] |
| | CPMF400 | 79.20 | — | 5 | Manual squeezing | — | 1 | [69] |
| | PDMS/CNF-4 | 70 | 2 g/g | 1.5 | Manual squeezing | 10 | 1 | [70] |
| | PDMS@WCS-20 | 75 | $9.84 \times 10^5$ g/m$^3$ | 10 | Mechanical compression | 6 | 1.5 | [64] |
| | OTS-rGO-WS | 88 | 7.28 g/g | — | mechanical compression | 10 | 1 | [11] |
| | CNT/PDMS-PU-4 | 88 | 20 g/g | 15 | Manual squeezing | 5 | 1 | [71] |
| MXene materials | MXene aerogel | 65.80 | 24.50 g/g | 35 | Mechanical compression | 5 | 0.8 | [61] |
| Carbon-based materials | CNT/RGO-1 sphere | 88 | 267 g/g | 10 | *n*-Hexane wash | 5 | 1 | [65] |
| | CNT/RGO monolith | 91 | 125 g/g | 15 | — | — | 1 | [65] |
| | RGO monolith | 78 | 105 g/g | 15 | — | — | 1 | [65] |
| | MF/rGO | 89 | — | 1.5 | Manual squeezing | 500 | 1 | [72] |
| | Carbonized Fabrics | 34.4 | — | — | — | — | 1 | [73] |
| | CNT/wood aerogel | 70 | 23.1 g/g | 10 | Manual squeezing | 10 | 1 | [74] |
| | rGO@PPS | 72 | — | — | — | — | 1 | [75] |
| | CR-S | 60 | 60 g/g | 10 | Manual squeezing | 7 | 1 | [76] |

## 5 Performance comparison of photothermal materials toward removal of oil spills

In recent years, the efficient and pollution-free application of photothermal materials in crude oil adsorption by converting solar energy into heat has attracted considerable attention. Photothermal materials in high-viscosity oil spill remediation rely on their unique structural properties, such as surface hydrophobicity, high surface area, and tunable pore structure. Judging the advantages and disadvantages of photothermal materials in the field of crude oil adsorption mainly depends on the adsorption capacity, adsorption time, cycle stability, and photothermal conversion performance. Table 1 summarizes the photothermal materials in crude oil adsorption in the past 5 years.

## 6 Summary and outlook

Photothermal conversion materials based on sunlight utilization technology can absorb sunlight, convert it into heat energy, and then reduce the viscosity of crude oil through the transfer of heat energy to achieve the purpose of crude oil adsorption. Metal materials have problems, such as complex preparation process, environmental pollution, high cost, and low photothermal conversion efficiency, limiting their application in crude oil adsorption. However, semiconductor materials are expensive, and some are difficult to prepare on a large scale. Long-term service may lead to reduced stability, which limits their practical application. Organic polymers are usually compounded with commercial sponges. Generally, their slow heating rate, low thermal conductivity, and low maximum equilibrium temperature limit their application in crude oil recovery.

In contrast, carbon-based materials have attracted much attention in crude oil adsorption due to their broad-spectrum absorption, tunable pore structure, stable mechanical properties, acid and alkali corrosion resistance, and high photothermal conversion efficiency. At present, the light absorption properties of most materials reach more than 90% and should no longer be the focus of research. The relationship between the physicochemical properties of materials and the photothermal conversion needs to be further explored to improve the energy conversion efficiency. Carbon-based materials are prone to defects, resulting in additional thermal resistance, which cannot exert their inherent excellent physical properties well. In addition, the extra-loaded photothermal absorption element is easily detached under long-cycle conditions, thereby significantly reducing the photothermal conversion effect, and further research is needed.

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