Supporting Information

A super-hydrophilic NH₂-MIL-125 composite film with dopamine-modified graphene oxide is used for water treatment

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The preparation of the GO

The modified Hummers method was used to prepare go. The specific steps are as follows: weigh 3 g graphite powder and add it to 1000 mL three-way flask, transfer the flask to ice water bath, slowly add 60 mL H_2SO_4 and 6 mL H_3PO_4 mixture to it, and ultrasonic until uniform.Secondly, slowly add 3g $NaNO_3$ and 15g KMnO4 through mechanical stirring, continuous stirring for one hour, transfer to the oil bath, under the condition of 35°C continuous stirring 2h. After stirring, 90 mL pure water was added and stirred for 2 hours at 95°C. In this process, the mixed solution gradually turned bright yellow and accompanied by bubbles.Then, add H_2O_2 aqueous solution (30wt%,150 mL including 70 mL H_2O_2) to a three-mouth flask. After 1h, add 7 mL dilute HCl solution (5wt%,50 mL) to react for 0.5h, and centrifuge the mixture of ethanol and deionized water until pH is neutral. The final product was ultrasonic for 4h and dried to obtain flake go, which was ground into powder for use.

Preparation of NH_2-MIL-125(Ti) nanomaterials

NH_2-MIL-125(Ti) was synthesized by solvothermal method.First, 0.56g of 2-amino-terephthalic acid was added into the mixed solution of dimethylformamide and methanol (DMF: MeOH=9:1) and treated with ultrasound for several minutes.Secondly, 0.2ml tetra butyl titanate was added to the solution, along with 0.3 mL acetic acid (to inhibit the hydrolysis of tetra butyl titanate). The solution was transferred again to a 50mL PTFE liner again and maintain at 150°C for 24 hours. The product was washed several times with DMF and methanol, and then dried overnight in an oven at 60°C to obtain the purified product.

Table S1 composition of composite film

Type of film	RGO(ml)	NH_2-MIL-125（Ti）(mg)
M0	4	0
M1	2	1
M2	2	2
M3	2	3
M4	2	4

Characterization

The crystal structure of materials was characterized by X-ray diffraction (XRD, PANalytical, Netherlands). The chemical composition and elements on the composite films were characterized by X-ray photoelectron spectroscopy (XPS, Escalab 250 spectrometer Walttham Inc., USA). The Fourier transform infrared spectroscopy (FTIR, WQF-520, Beijing Rayleigh analytical instrument Company, China) were used to characterize chemical composition of materials. The surface morphology and roughness of the composite films were measured by atomic force microscope (AFM, SPA300HV, Japan). The appearance of composite film were characterized by scanning electron microscope（SEM JSM-7500F, JEOL, Japan）. The hydrophilicity of the membrane was measured by an integrated measurement system of interface parameters (Beijing Hake, XED-SPJ). And the absorbance of dyes was measured by UV–vis-NIR spectrophotometry on UV-762 (Shanghai Precision Scientific Instrument Company). The water environment carbon and nitrogen comprehensive analyzer (TOC, SHIMADZU, Japan Shimadzu) was used to determine the oil content in the feed and permeate.

Separation and filtration evaluation

Vacuum filtration device was used to study the removal effect of the composite film on different oil-in-water emulsions and dyes. Before the filtration experiment, the newly prepared complex film was placed on the vacuum filter device at a pressure of 0.09 MPa and pre-pressed with deionized water for 10 min to ensure the tightness of the filtration system, and the effective area of the middle filter layer of the filter device was 12.56cm^2.

Flux calculation of the film is as follows (1):

$$J_w = \frac{V}{A \times t}$$

(1)

Wherein: J_w-flux, L/(m^2·h);

V - Volume of pure water permeable to the film, L;

A - Effective film area, m^2;

t - Filtration time, h.

The retention rate of the film represents the ability of the film to separate pollutants, and the interception rate of the film is directly proportional to the separation effect of the film. In this experiment, the concentration of oil in the simulated wastewater used for filtration was 20 mg L^{-1}. After pre-pressing the newly prepared film for 10min, measure the pure water once and pour the bottom solution into the designated wastewater area, and then pour the simulated wastewater solution in the 20 mL configuration into the filter unit for separation experiments. After the solution filtration is completed, filtrate is collected and measured by the water environment carbon and nitrogen comprehensive analyzer, and the interception rate of each pollutant by the film is calculated according to the standard curve. Calculations such as (2):

$$R = \left\{1 - \frac{C_p}{C_f}\right\} \times 100\%$$

(2)

Where: R - the retention rate of the contaminant, %;

C_p - initial concentration of contaminants, mg·L^{-1};

C_f − the concentration of pollutant after filtration, mg·L^{-1}.

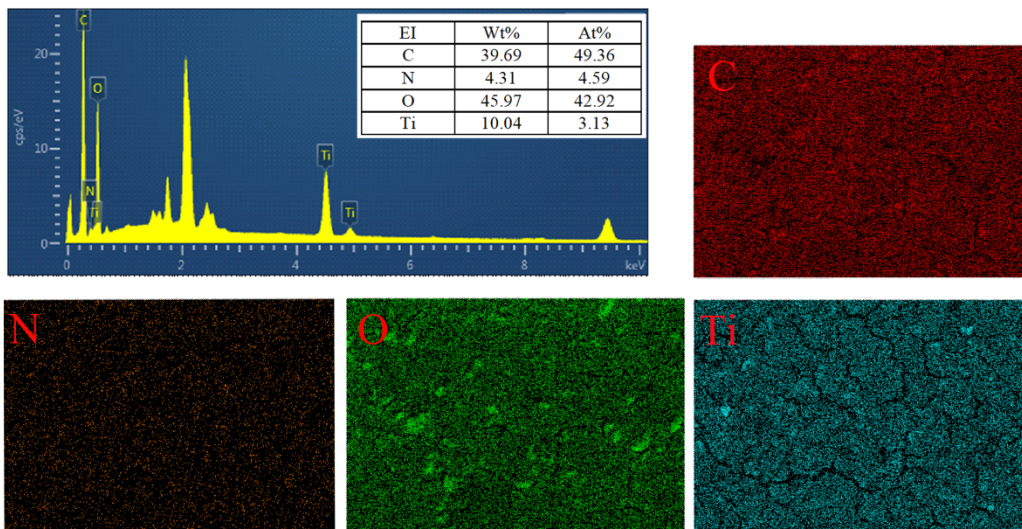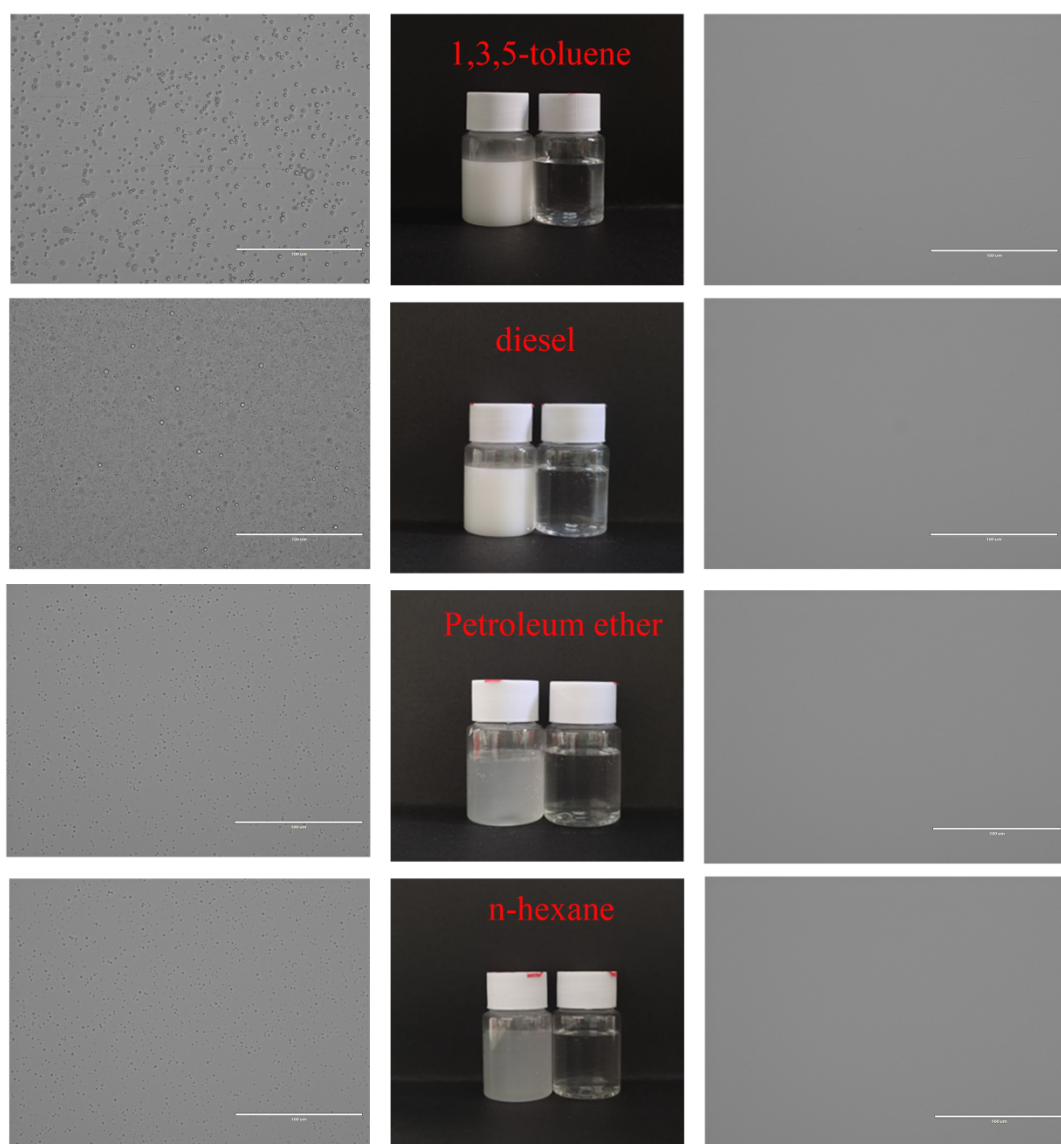| EI	Wt%	At%
C	39.69	49.36
N	4.31	4.59
O	45.97	42.92
Ti	10.04	3.13

Fig. S1 EDS and element distribution of M3

Fig. S2 Optical microscope images of M3 before and after various oil-in-water emulsions.
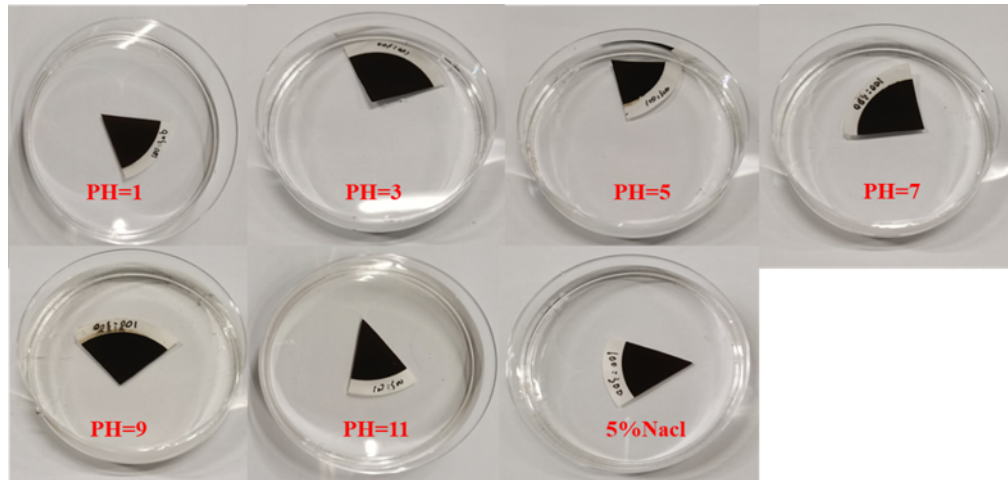
Film stability testing

Fig. S3 Photos of the membrane soaked in different acid and base conditions for 30 days