

Supplementary Information:

One-step fabricated wettable gradient surface for controlled directional underwater oil-droplet transport

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Experimental section:

Preparation of gradient graphite plate: A kind of high density graphite plate (rinsed with deionized water and ethanol successively before use) with the size of $30 \times 10 \times 2$ mm manufactured by Ji Xing Sheng An Co. (Beijing, China) was perpendicularly immersed into 0.05 M electrolyte solution sodium hydrate (NaOH) and treated via a one-step anodic oxidation method. A narrow-strip copper plate as the cathode facing the bottom of graphite plate was used to form the current gradient. The electrochemical process was performed at a constant current between 0.3 A and 0.6 A. In addition, a valve attached to the bottom of the chamber was used to discharge the electrolyte solution gradually from the reaction vessel to control the oxidation time, which is consistent with the effect of oxidation current gradient (see Fig. S1). With the cooperation of the two effects above, the wettable gradient could be adjusted easily. After anodic oxidation, the graphite plate was rinsed thoroughly with freshly deionized water and dried at room temperature under vacuum.

Characterization: Water contact angles (WCAs), oil (chloroform) contact angles under deionized water (UOCAs) and oil contact angles in air (OCA) were measured by the optical contact angle meter system (OCA40Micro, Dataphysics Instruments GmbH, Germany). A drop of deionized water or analytically pure oil under deionized water was dripped onto the samples and the static contact angle (CA) was determined by the average of at least five measurements. The dynamic transport process of underwater oil droplet was also recorded by the system. Time zero was chosen to be the frame in which deposited droplets contacted with graphite plate. Scanning electron microscopy (SEM) equipped with energy dispersive spectrometer (EDS) (JSM-6500F, JEOL) was employed for

the microstructures and chemical composition identification of graphite plate surface. The adhesion force was measured by surface interface tensiometer (DCA21, Dataphysics, Germany).

Supplementary Figure Legend: Fig. S1-S7

Fig. S1:

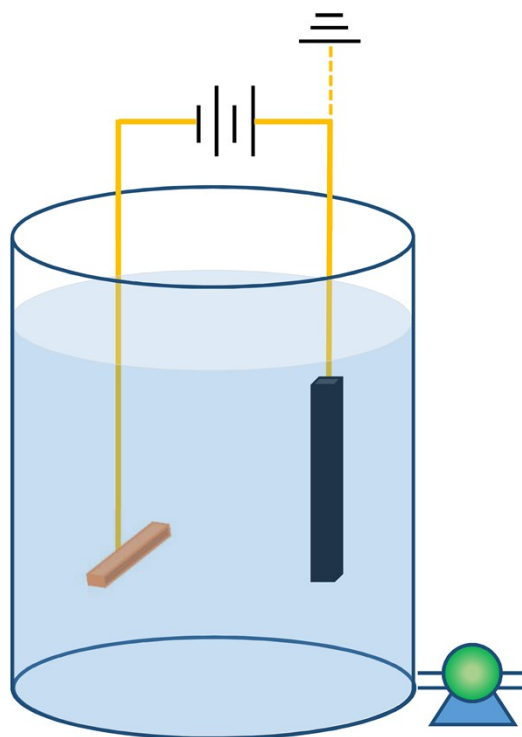


Fig. S1 Apparatus for anodic oxidation

Fig. S2:

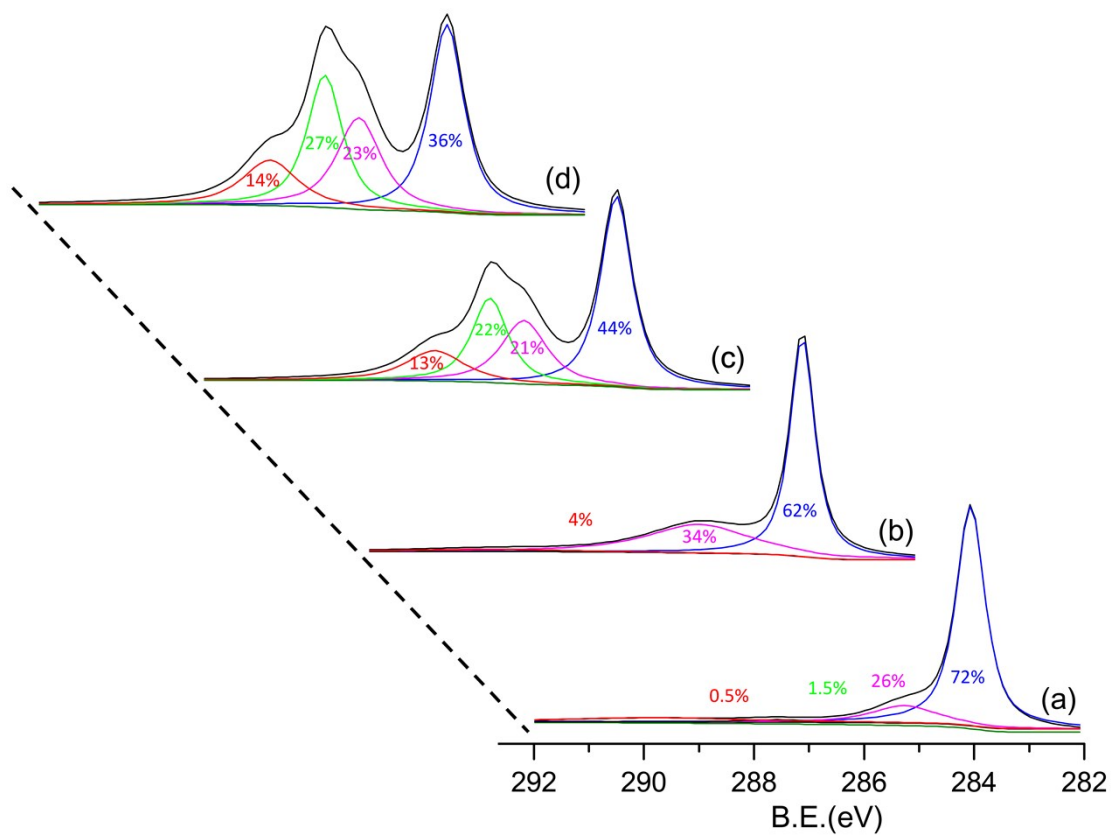


Fig. S2 High-resolution XPS C 1s spectra at different areas on oxidized graphite plate: (a) 0 mm, (b) 10 mm, (c) 20 mm, (d) 30 mm (from T part to B part). Clearly, the increase of the O 1s /C 1s ratios attributes to the increase of carboxyl and carbonyl contents.

Fig. S3:

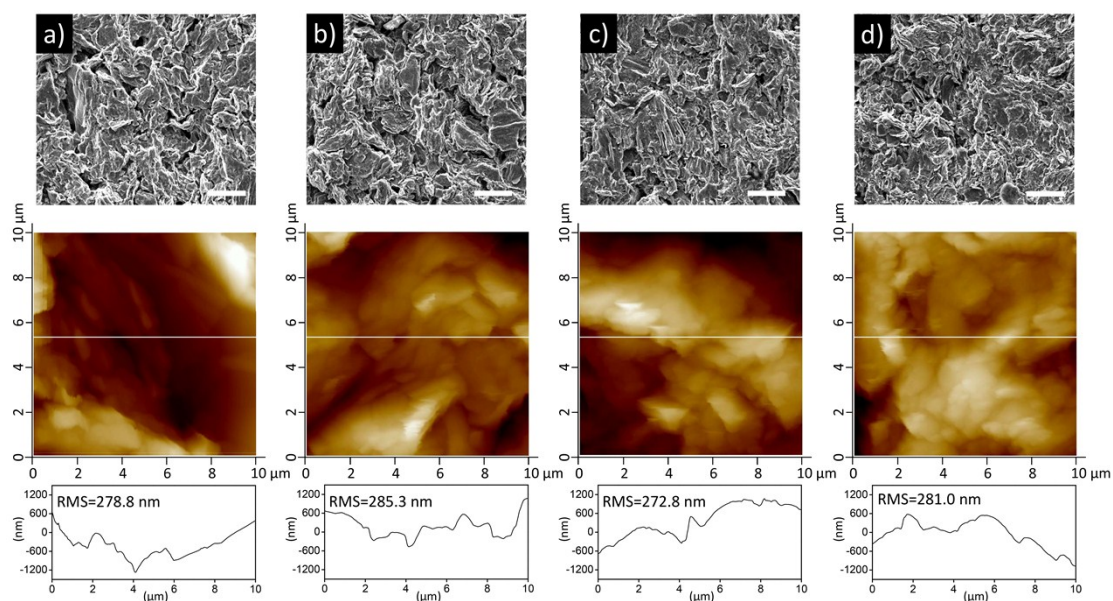


Fig. S3 SEM and corresponding AFM images of AO graphite plate surface at different areas. (a) 0 mm, (b) 5 mm, (c) 15 mm, (d) 25 mm (from T part to B part). Scale bars, 25 μm. It's clear that there are many micropores on the graphite plate surface and porous structures are observed. The root-mean-square (RMS) roughness of the sites are 278.8 nm, 285.3 nm, 272.8 nm and 281.0 nm, respectively. The images of different areas of the AO surface show little difference between each other.

Fig. S4:

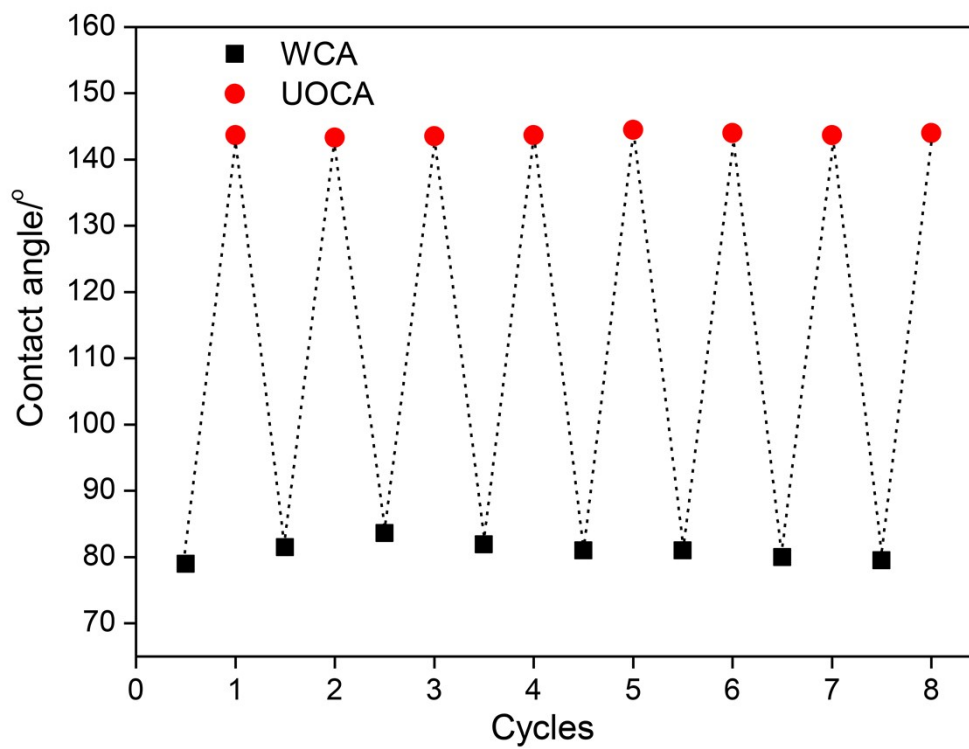


Fig. S4 Repeated cycling measurement of WCA and UOCA on the AO graphite plate surface indicates excellent reproducibility and stability.

Fig. S5:

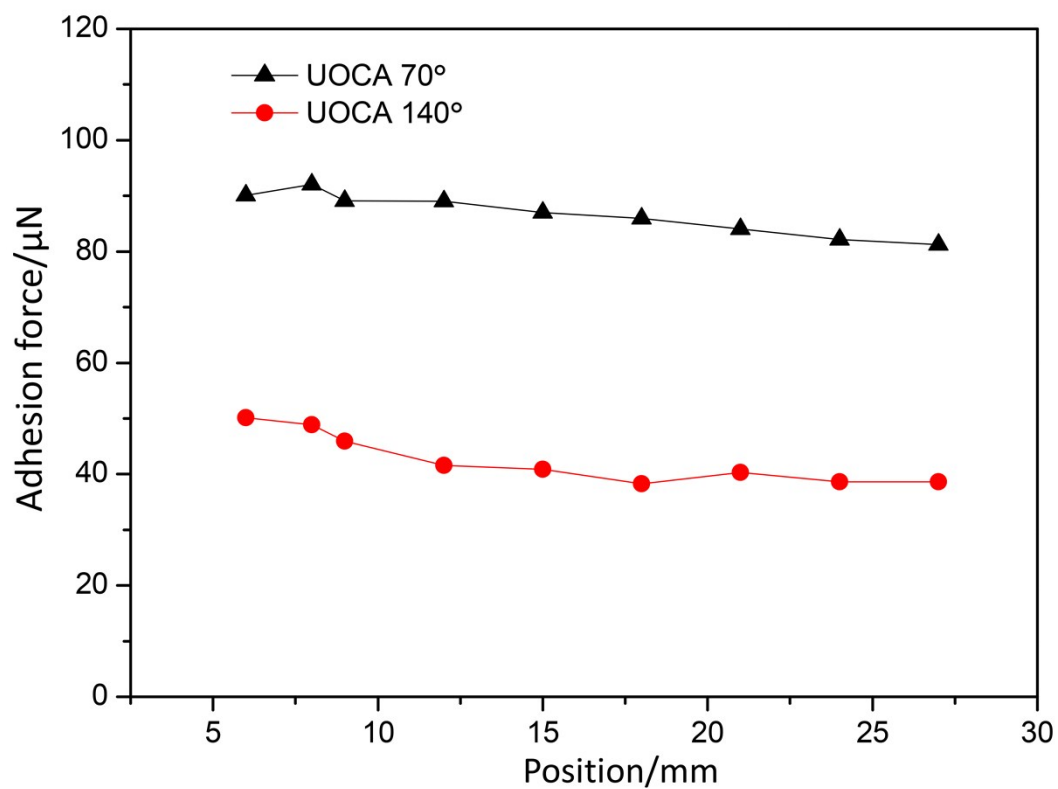


Fig. S5 The adhesion forces of underwater oil on the untreated graphite plate surface with a UOCA 70° and the AO treated graphite plate surface with a UOCA 140°.

Fig. S6:

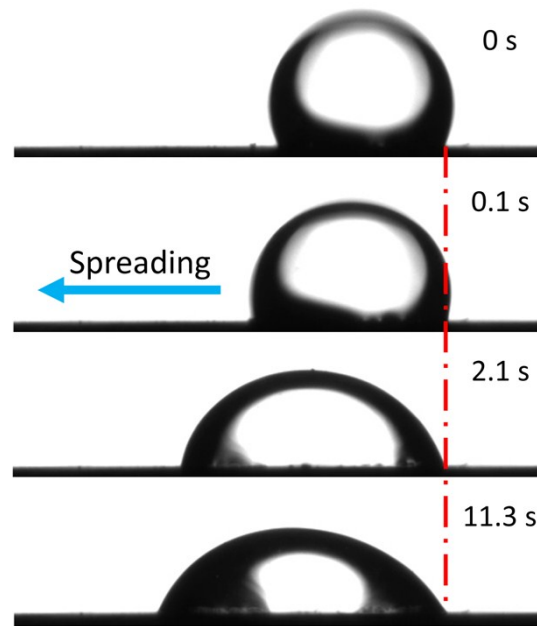


Fig. S6 The spreading process of an underwater oil droplet on the AO surface with an underwater oil wettable gradient $23^\circ/\text{mm}$. No apparent movement shows up due to high adhesion. The volume of the oil droplet is $10 \mu\text{L}$.

Fig. S7:

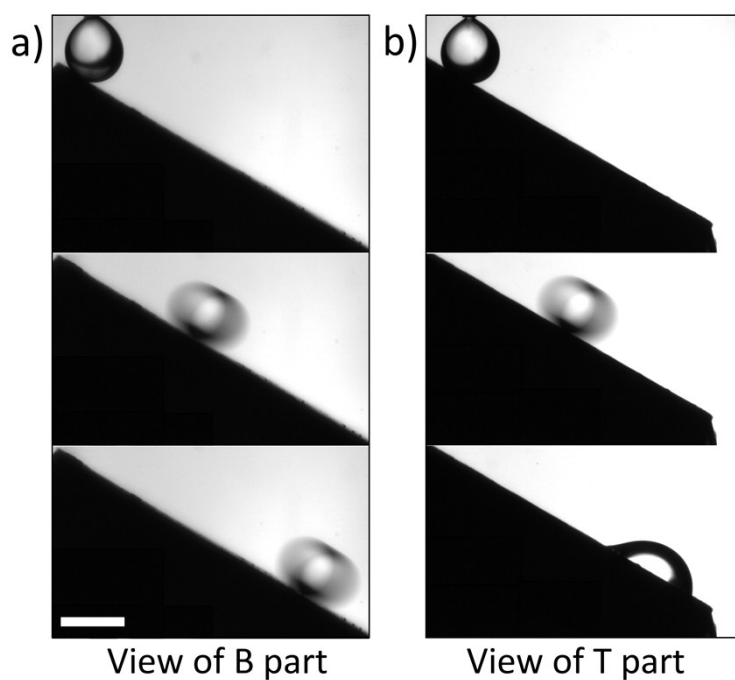


Fig. S7 Movement behaviours of underwater oil droplets on Sample-D at tilted angle 30° . The scale bar is 2 mm. The underwater oil droplets totally roll off the surface at this tilted angle. Because of the limit of the camera scope, only the movement behaviours at B part (a) and T part (b) are displayed. Here, it is observed that the underwater oil droplet dripped at B part rolls off the surface and rolls out of sight. As for the oil droplet dripped near T part, it rolls off the surface and is finally pinned at the end of the surface due to large adhesion force, where is not oxidized.