Computational Methods

1. Heat-transfer simulations. To gain more insight into the thermal transmission and energy utilization of cleanup viscous crude oil in our experiments, heat-transfer simulations were performed with the finite element method (FEM). In all simulations, the thermal transmission is described by the classical heat-transfer model:

\[ \rho C_p \frac{\partial T}{\partial t} = \nabla \cdot (k \nabla T) + Q \]  

(1)

where \( T \), \( \rho \), \( C_p \), \( k \), and \( Q \) denote the temperature, mass density, heat capacity, heat conductivity, and heat source, respectively. Convective heat flux boundaries were imposed on the outer surfaces of our models. The convective heat flux is driven by the temperature difference between the outer surface and the surrounding ambient:

\[ q = -h(T_{\text{sur}} - T_{\text{ext}}) \]  

(2)

where \( q \) and \( h \) denote the inward heat flux and the heat-transfer coefficient, respectively. \( T_{\text{sur}} \) and \( T_{\text{ext}} \) represent the surface and external temperatures. In the oil cleanup FEM model as showed in Supplementary Fig. 12a, the upper bulk denotes the cubic graphene wrapped sponge (GWS, 20 mm \( \times \) 20 mm \( \times \) 20 mm). The bottom part is bulk water with 100 mm of diameter and 50 mm of depth.
The brown component represents the crude oil with 60 mm of diameter and 5 mm of thickness. The marked red region with a height of \( x \) is the heated source, corresponding to the joule-heated part in the GWS in our experiments. In the simulations, the initial temperature of all components are 24 °C (ambient temperature in the experiment). The power density of the heat source is defined as \( n \) W/cm\(^3\). Here, the value of the power density \( n \) is related to the height \( x \) of the heated region in FEM simulations, which satisfies the rule that the maximum temperature of the heated region can reach 250 °C. The quantity of heat transferring into oil or water was calculated by the integral:

\[
\Delta Q = \iiint (T - T_0) C_p \rho dV
\]

where \( T_0 \) and \( V \) are initial temperature and the volume of the oil or water, respectively. The quantity of heat transferring into the air was computed by:

\[
\Delta Q_{air} = \int_i \left( \iiint_A -q dA \right) dt
\]

where \( A \) is the area of the outer surface. Then, we defined the ratio \( \eta \) to characterize the energy utilization in the cleanup oil process:

\[
\eta_i = \frac{\Delta Q_i}{E_{total}} = \frac{\Delta Q_i}{n Vi}; \quad i = \text{oil, water, or air}.
\]

2. Electro-thermal coupling simulations. GWS can be heated by electric current because of the electro-conductibility of internal coated graphene. In order to investigate the influence of electrode size and conductive region on temperature distribution in GWS, extensive electro-thermal coupling simulations were performed. Sketch map of GWS and GWS-MS model in thermoelectric coupling simulations are showed in Fig. 4a. In the simulations, two electrodes (height \( x \)) were located at two opposite sides with an electric potential difference. The power density generated by electric current is given by:

\[
Q = J \cdot (-\nabla V)
\]

where \( J \) and \( V \) denote current density and electric potential (voltage), respectively. In this electro-thermal coupling problem, the electric current distribution is solved by the Maxwell equations, then the heat generated by electric current is regarded as the inward heat source to solve the temperature distribution based on classical heat-transfer model. Convective heat flux boundaries were applied on all outer surfaces. The electrode size (height \( x \) in Fig. 4a) was a controllable
variable in the FEM simulations. The imposed electric power was fixed to 0.423W (corresponding to the input electric power density 0.094 W/cm$^3$ in the experiment, Fig. 4b) for different electrode sizes.

3. Cleanup-oil simulation. Furthermore, we simulated the dynamic process of cleanup-oil to investigate the thermal diffusion (Supplementary Fig. 13). In the simulation model, bottom-half of the cubic sponge was treated as uniformly distributed heat source with the power of 1.6 W corresponding to the electrically conductive GWS. The thickness of the oil part was set to be 5 mm. Convective heat flux boundaries was applied on all outer surfaces. The deformable geometry dynamic boundary was imposed to simulate the sinking process of the sponge. First, part of sponge was initially immersed in the oil with the depth of ~1 mm for 20 seconds. Then, the sponge submerged into oil under a given sinking velocity of ~0.045 mm/s in this simulation, until that the bottom of the sponge was closed to the oil-water interface. In the submerging process, the part of immersed sponge was regarded as in a full of oil state, and the material property of this part was changed accordingly. All the simulation parameters (Table S1) were referred to the measured values in the experiments. In this dynamic process, temperature distribution of the vertical cross-section was tracked in the simulation (Supplementary Movie 4). The percent of heat transferred into different components were calculated in the entire submerging process.

Table S1. Material properties used in simulations

<table>
<thead>
<tr>
<th>Sample name</th>
<th>Thermal conductivity W/(m K)</th>
<th>Heat capacity J/(Kg K)</th>
<th>Density Kg/m$^3$</th>
<th>Heat transfer coefficient W/(m$^2$ K)</th>
<th>Electrical conductivity S/m</th>
</tr>
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<tbody>
<tr>
<td>GWS</td>
<td>0.02558</td>
<td>3670</td>
<td>10</td>
<td>12</td>
<td>0.28</td>
</tr>
<tr>
<td>MS</td>
<td>0.03326</td>
<td>2380</td>
<td>10</td>
<td>12</td>
<td>-</td>
</tr>
<tr>
<td>Oil</td>
<td>0.148</td>
<td>1770</td>
<td>940</td>
<td>2</td>
<td>-</td>
</tr>
<tr>
<td>Water</td>
<td>0.55</td>
<td>4200</td>
<td>1000</td>
<td>5</td>
<td>-</td>
</tr>
<tr>
<td>Electrode (Silver)</td>
<td>429</td>
<td>55</td>
<td>10600</td>
<td>5</td>
<td>$6.3 \times 10^7$</td>
</tr>
<tr>
<td>Oil soaked GWS</td>
<td>0.1537</td>
<td>1596</td>
<td>920</td>
<td>-</td>
<td>0.28</td>
</tr>
</tbody>
</table>
Supplementary Figure 1 | (a) SEM image of the melamine sponge (MS)/GO composite which was obtained by directly drying the melamine sponge full filled with GO solution (3 mg/mL) under 65 °C. It clearly shows that the majority of the pores of the sponge were blocked by the GO film. (b-f) SEM images of the MS/GO composites which were obtained by centrifuging the melamine sponges full filled with GO solution (3 mg/mL) at 500 rpm, 1000 rpm, 2000 rpm, 3000 rpm, and 4000 rpm, respectively. These SEM images shows that the number of pores in the sponge that blocked by GO film decreased with the increase of the centrifugation speed, and negligible GO film appeared in the pores when the centrifugation speed increased up to 3000 rpm. (g) Conductivities of the reduced MS/GO composites mentioned in a-f (all the samples were reduced by HI). When the centrifugation speed increased over 1000 rpm, the conductivity of the reduced MS/GO composites almost kept constant, which indicates that the increased centrifugation speed had negligible effect on the amount of GO nanosheets on the skeletons of the sponge. Therefore, the centrifugation process removed the GO solution in the pores while leaving a small amount of GO solution on the skeleton of the sponge, which plays an important role in obtaining three dimensional RGO conductive networks without blocking the pores of the sponge.
Supplementary Figure 2 | (a) Optical image of MS (left), MS@GO (middle), MS@RGO (right). (b) and (d) SEM images of MS. High magnification image shows a MS skeleton with smooth surface. (c) and (e) SEM images of MS@RGO. High magnification image obviously shows a lot of wrinkles on the skeleton, which indicates a thin layer RGO film coated on the MS skeleton. (f) Raman spectrums of MS, MS@GO and MS@RGO. D peak (1356 cm$^{-1}$) and G peak (1594 cm$^{-1}$) are the feature peaks of GO and RGO, and the increase D/G intensity ratio demonstrated the reduction of GO$^1$. 

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**Supplementary Figure 3** | (a) Optical image of MW (left), MW@GO (middle), MW@RGO (right). (b) and (d) SEM images of MW. High magnification image shows a MW fiber with smooth surface. (c) and (e) SEM images of MW@RGO. High magnification image shows obvious wrinkles that indicates the existence of RGO coating wrapped around MW fiber. (f) Raman spectrums of MW, MW@GO and MW@RGO, demonstrating the GO coating and reduction of GO to RGO.
Supplementary Figure 4 | (a-d) Photo images of the original melamine sponge (10 × 30 × 2 cm³), GO solution soaked melamine sponge, GO coated melamine sponge, RGO coated sponge, respectively. (e) A piece of GO solution soaked melamine sponge (10 × 30 × 2 cm³) was mounted in the drum of an industrial centrifuge. (f) During the centrifuge process, the GO solution was removed from the sponge and recycled. (g) Side view of the RGO coated melamine sponge (10 × 30 × 2 cm³). (h, i) Photo images of a melamine sponge which was partially coated with GO, and corresponding reduced one. We named the partially coated sponge as GWS-MS. The un-coated region was modified with octadecyltrichlorosilane to realize hydrophobicity.

Supplementary Figure 5 | Weight loss of MW in air versus temperature. When the MW was heat up to 600 °C (air atmosphere) which is close to the breakdown temperature of graphene, only 1.11% of weight loss was observed.
Supplementary Figure 6  | (a) Schematic illustration of the setup to measure the surface temperature of the MW@RGO with different applied voltages. The two ends of the MW@RGO monolith were clamped by two carbon rods which served as the electrodes due to their stability under high temperature. Multivoltage steps were applied on the MW@RGO monolith. Before applying the next step voltage, the former voltage was turned off in advance to let the MW@RGO monolith cool to room temperature. The surface temperatures of MW@RGO were measured by a thermal IR camera (InfraTec, Vario CAM hr head 640) and corresponding current densities were recorded by a digital multimeter (Agilent, 34411A). From the thermal IR images, maximum and average temperatures of the sponge surface were obtained. The resistance of MW@RGO monolith was calculated according to Ohm’s law \( R = \frac{U}{I} \). (b) Photography of the experiment setup for the measurement of surface temperature and resistance of MW@RGO applied with different voltages.
Supplementary Figure 7 | The plots of resistance and average surface temperature (AST) of MW@RGO applied with 43 V as a function of time. It should be noticed that no obvious change in both temperature and resistance was observed when MW@RGO was heated for 13.5 h. (Inset) IR Thermal images of MW@RGO after heated for 0 h, 4.5 h, 9 h and 13.5 h, respectively. The size of MW@RGO between the two carbon rods is 2 cm × 1.2 cm × 0.5 cm.
Supplementary Figure 8 | (a) Schematic illustration of the customized setup for the wicking method\(^2-4\) (left). A MS@RGO monolith (2 cm × 2 cm × 1.5 cm) was fixed below the force sensor of a universal mechanical testing machine (Instron 5565A) by four iron wires. Two side walls of MS@RGO (opposite position) were coated with silver paste (Dupont 4929N) as the electrodes and connected to the power supply through copper wires. The viscous crude oil was put on a vertically moveable stage which can push the oil surface up to contact with the bottom surface of MS@RGO. The oil can be heated up and maintained a certain temperature by placing a hot plate beneath the oil vessel. Right figure is the photography of the real setup. (b) Schematic illustration of the whole measurement processes. Firstly, the stage was gradually lifted up to get the oil surface contacted with the bottom surface of MS@RGO and fixed at this position. At the moment of touching the bottom surface of MS@RGO with the oil surface, the sorption process began. At this time, the weight of MS@RGO jumped to a certain value \(W_{t1}\), which was caused by the surface tension of the oil. Then, the weight of MS@RGO increased gradually by the absorption of the oil into MS@RGO. After reaching the saturated oil sorption capacity of MS@RGO, the stage was moved down to separate the bottom of MS@RGO from the oil surface. During this operation, weight increased sharply again at first due to the surface tension of the oil and then the total weight of MS@RGO decreased to a certain value of \(W_s\), which was the final weight of heavy oil absorbed by the MS@RGO. To eliminate the surface tension effect and clearly show the real oil absorption kinetics, the oil sorption curve (dashed line in Supplementary Fig. 8b) was obtained by deducting \(W_{t1}\) value from the original data. Finally, \(K_s\) could be derived from the slope of the linear relation between the mass of oil absorbed per unit area \(m_t\) and the square root of absorption time \((t^{1/2})\).
**Supplementary Figure 9** | (a) Schematic of the measurement of surface temperature of GWS. (b, c) With the input electric power of 0.27 \( \text{W/cm}^3 \) (20 V) and 0.58 \( \text{W/cm}^3 \) (30 V), the average surface temperatures of the bottom surfaces of GWS were 97.54 °C and 148.24 °C respectively.

**Supplementary Figure 10** | Clean up viscous crude-oil spills on sea water by MW@RGO without (a) and with (b) the aid of joule heating. The average surface temperature of MW@RGO was 250 °C. Room temperature was 25 °C.

**Supplementary Figure 11** | Schematic illustration of the oil cleanup process by joule heated GWS. In the real scenario of the oil spill clean-up process, the GWS was standing on the oil surface at the initial stage due to the light weight of GWS and the high viscosity of the crude oil. Then, with the aid of joule heat to lower the viscosity of the crude oil, the crude oil was gradually absorbed into GWS. Meanwhile, the GWS gradually move down until it reached the water surface.
Supplementary Figure 12 | (a) Schematic illustration of the oil-cleanup model in simulation. Simulated ratio of heat that transferred to air (b) and water (c). Simulated ratio of heat transferred to water and air (d), and oil (e) during the joule heating process. This simulation was done under the consumption that GWS keep it original position, and oil is not absorbed into GWS. $x$ is the thickness of the heating region. The size of GWS is 2 cm $\times$ 2 cm $\times$ 2 cm. The amount of heat transferred to air decreases with the decrease of heating region (Supplementary Fig. 12b), while that to water increase with the decrease of heating region (Supplementary Fig. 12c). The combined results are shown in Supplementary Fig. 12d, the total quantity of heat transferred to water and air decreases with the decrease of heating region. Meanwhile, the heat transferred to oil increases when the heating region is concentrated (Supplementary Fig. 12e).
**Supplementary Figure 13** | The displacement change of GWS-MS-10-Pmax as a function of oil absorbing time. At the initial 20 s, although the displacement of GWS-MS-10-Pmax increased to ~1 mm quickly due to the capillary force, the oil sorption speed was slow. After 20 s, the sponge submerged into oil at sinking velocity of ~0.045 mm/s. In the simulation (Computational Method 3), we let GWS-MS-10-Pmax heat up at the displacement of 1 mm for 20 s, then sinking into oil at the sinking velocity of 0.045 mm/s.

**Supplementary Figure 14** | (a) Schematic of the positions where the temperature change were calculated. (b) Calculated temperature change of the positions shown in (a). (c) Viscosity change of the crude oil versus temperature. (d) Displacement change of GWS-MS-10 without joule heating versus oil absorbing time. The whole oil sorption time is 1994 s. GWS-MS-10-Pmax (107 s) saved the oil sorption time by 94.6%.
Supplementary Figure 15 | The sinking speeds of GWS-MS-10 with different area under the same electric power density (0.098 W/cm³). Room temperature 26 °C.

Supplementary Figure 16 | (a) Schematic of GWS which is connected with two copper wires. (b) Optical image of GWS which is connected with two copper wires. (c) Thermal image of the GWS with two electrodes (left) and optical image of the power supply (right, 15 V, 0.015 A). (d) Schematic of GWS which is connected with multiple crossed copper wires. (e) Optical image of GWS which is connected with multiple crossed copper wires. (f) Thermal image of the GWS with multiple electrodes (left) and optical image of the power supply (right, 15 V, 1.778 A).
Supplementary Figure 17 | (a) Schematic of the pillar-array electrode design for large scale GWS. (b) Photo image of the pillar-array electrode. (c) Thermal image of GWS-MS (30 cm × 10 cm × 2 cm) under 0.016 W/cm³ of power input. MST is 207.02 °C, AST is 125.01 °C. (d-f) A piece of GWS-MS (30 cm × 10 cm × 2 cm) was immobilized on the pillar-array electrode for oil absorption test. 199 g of crude oil was poured on the water surface of glass container (area of 11.2 cm × 30.6 cm). Room temperature was 22 °C. The oil thickness was ~ 6 mm. The GWS-MS was applied with electricity power of 0.016 W/cm³ (42.2 V). After 4 min of oil absorption, the viscosity crude-oil spill was absorbed into the GWS-MS.

Supplementary Figure 18 | Schematic illustration of the compression recovery mode. A MS@RGO monolith (2 cm × 2 cm × 1.5 cm) was firstly filled with viscous crude oil and then fixed to a parallel-jaw vice. The MS@RGO was compressed to 75% strain and the oil was collected by a petri dish.
**Supplementary Figure 19 |** (a) Schematic illustration of the in situ pumping recovery mode. 30 mL of sea water was add into a 100 mL beaker, then ~ 20 mL of crude oil was added to the beaker, forming viscous crude-oil spill. MS@RGO (2 × 2 × 2.9 cm³), whose two opposite side walls were coated with silver paste (in square loop, right figure) and connected to a power supply by copper wires. The MS@RGO was inserted to the water. Then one tip of a pipe was insert into the MS@RGO, and the other tip of the pipe was connected to the inlet of the self-priming pump. The pump’s outlet was connected to a collecting vessel by another pipe. The voltage applied on MS@RGO was 17 V, after heating for 1 min, started the pump with a voltage of 5.76 V. The pumping though unheated MS@RGO was also performed under the same pump voltage. Before pumping, the MS@RGO monoliths were all filled with crude oil. (b) Optical photography of the MS@RGO coated with silver electrodes. (c) Thermal IR image of MS@RGO applied with 13.72 V. The heat spots mainly located in the area between the silver electrodes, which contacted with the oil layer.

**Supplementary Figure 20 |** Plot of oil recovery percent for oil (at 25 °C, 75 °C respectively) inner MS@RGO as a function of time during the compressing process of MS@RGO (from 0 to 75% of compressive strain).
Supplementary Figure 21 | Plots of pressures applied on four MS@RGO monoliths (2 × 2 × 1 cm³) which filled with oil at 25 °C, 40 °C, 60 °C, and 90 °C, respectively as a function of compressive strain. A dramatically decrease of pressure was observed as the oil temperature increased.

Supplementary Figure 22 | (a) Cyclic compressive stress-strain curves of a MS@RGO monolith at a set strain of 75%. The compressive rate was 5% strain per second. The inset images show the sequential photographs of the MS@RGO monoliths during the compression process. Even though a little deformation of MS@RGO appeared after 1000 compress-release cycles, it still kept its elasticity well. (b) Oil absorption capacities of the MS@RGO monolith (1.91 cm × 1.82 cm × 1.82 cm) at each oil absorption-recovery cycle. The oil temperature was 95 °C. The compressive strain of each cycle was 80%. The oil sorption capacity of MS@RGO was not reduced after the oil absorption-recovery cycle test. The percentages of recovered oil from MS@RGO at each cycle were over 70%. (c) Resistance change of MS@RGO during the oil absorption-recovery cycle test. (d) SEM image of the MS@RGO after 10 oil absorption-recovery cycles. The dash circle regions show the broken fibers, which is the reason why the resistance of MS@RGO increased after the oil absorption-recovery cycle test.
**Supplementary Movies**

**Movie 1:** Surface temperature increase of MS@RGO under an applied voltage.

**Movie 2:** Permeating behavior of one oil droplet on the surface of MW@RGO with and without joule heating.

**Movie 3:** Clean-up of viscous crude-oil spill by MW@RGO with and without the aid of joule heating.

**Movie 4:** Simulation of the temperature change of joule heated GWS-MS-10 and oil during the oil absorption process.

**Movie 5:** Clean-up of crude-oil spill by a large-size GWS-MS-10 with the aid of joule heating.

**Movie 6:** Recovery of viscous crude oil (25 °C and 75 °C, respectively) from GWS by compressing GWS.

**Movie 7:** Continuous collection of the viscous crude oil from water surface by *in-situ* pumping through GWS with and without the aid of joule heating.

**References**


