

## Supporting Information

### **The alignment of thermally conducting nanotubes making high-performance light-driving motors**

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## Supplementary Notes

### 1. Preparation of aligned multi-walled carbon nanotube films

Spinnable multi-walled carbon nanotube (MWCNT) array was synthesized by chemical vapor deposition. In a typical synthesis, Fe (1.2 nm)/Al<sub>2</sub>O<sub>3</sub> (5 nm) on a silicon wafer was used as the catalyst, ethylene was used as carbon source with a flowing rate of 90 sccm, and a mixture of Ar (480 sccm) and H<sub>2</sub> (30 sccm) was used as the carrier gas. The growth was made at 750 °C, and the thickness of the vertically aligned MWCNT array was appropriately 200 μm. Aligned MWCNT sheets were directly drawn out of one sidewall of an MWCNT array. Aligned MWCNT films (thickness of ~ 800 nm) were obtained by stacking 40 MWCNT sheets together.

### 2. Photo-thermal conversion of aligned MWCNT and PDMS films

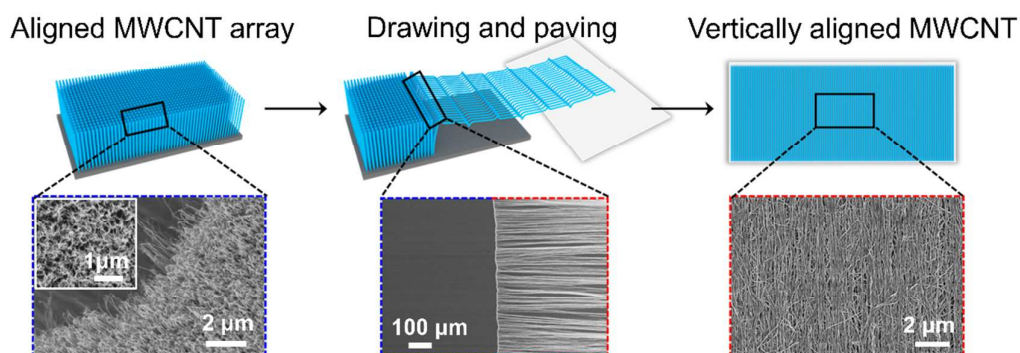
An aligned MWCNT film (2×2 cm<sup>2</sup>) and a spin-coated PDMS film (2×2 cm<sup>2</sup>) were irradiated under the same light source (500 mW/cm<sup>2</sup>) for 180 s. This test was first measured in air and then in water. Time-independent temperature variations were recorded and analyzed frame by frame of infrared pictures. Average temperatures of two films were extracted and demonstrated in Figure 3b.

### 3. Aligned nanostructure regulating thermal transmission

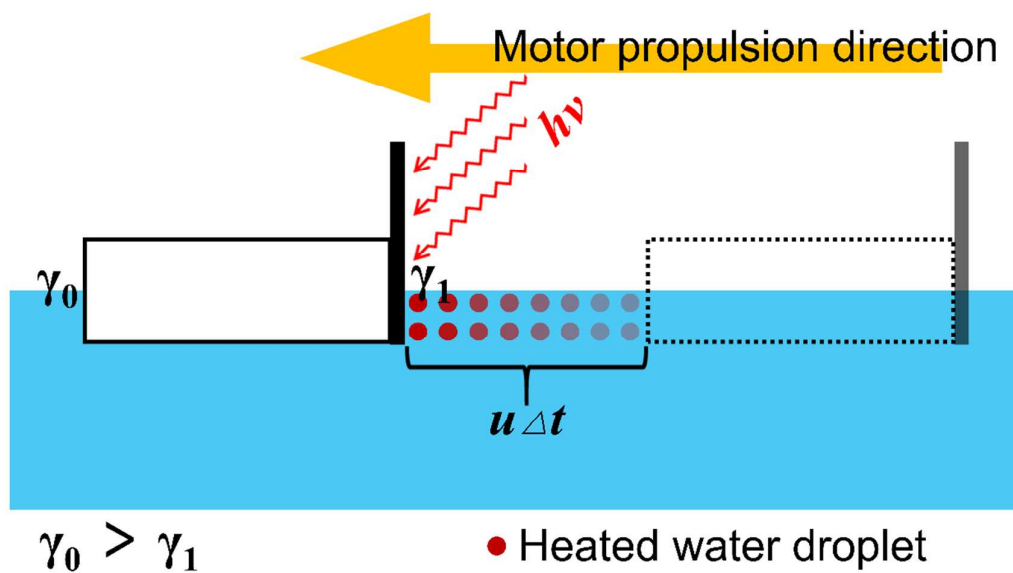
To probe the dependence of light-induced thermal transmission on aligned-MWCNT direction, the heat diffusion of two MWCNT films with the aligned MWCNTs being vertical and horizontal to the water surface were compared (testing setup in Figure S7, results in Figure S8). The top halves of the MWCNT films were heated by the same light source (power density of 100 mW/cm<sup>2</sup>) with the bottom halves being shielded from the light irradiation. The bottom halves were then investigated for the

temperature variation under light irradiation for 40 s. The vertically aligned MWCNT film was observed for a rapid temperature increase to 60.5 °C. In contrast, the bottom half of the horizontally aligned MWCNT film exhibited a much lower heating rate with most portion of the film below 40 °C. This phenomenon suggested that the thermally conducting MWCNTs favored a direct thermal transmission to the water surface for an enhanced light utilization.

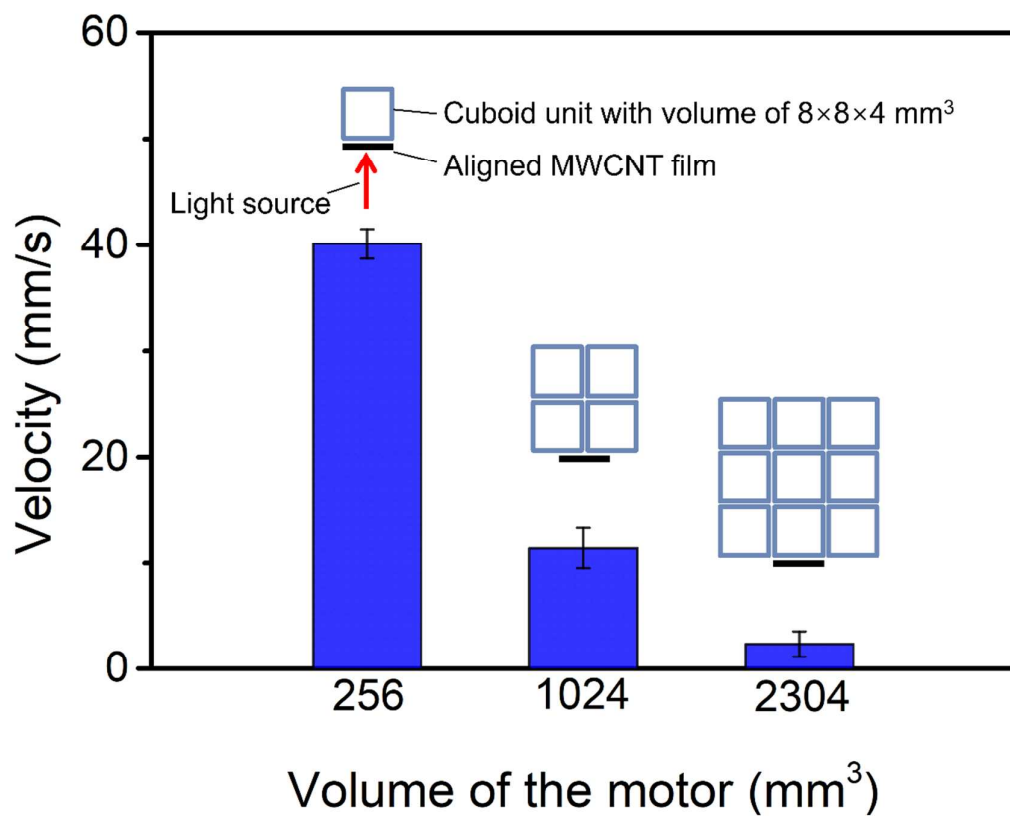
## Supplementary Figures



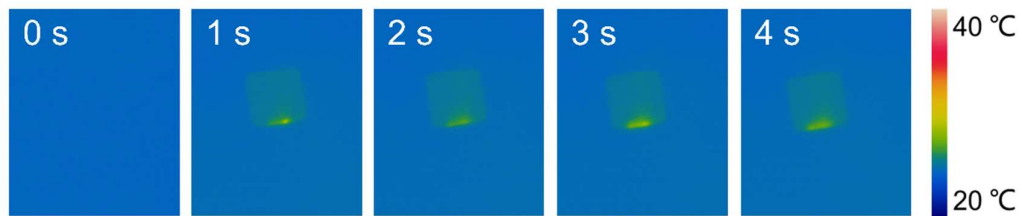
**Figure S1.** Aligned MWCNT array was synthesized by chemical vapor deposition. Aligned MWCNT sheets were directly drawn out of one sidewall of an MWCNT array. Aligned MWCNT films (thickness of  $\sim 800$  nm) were obtained by stacking layers of MWCNT sheets together on an ultrathin quartz slice. The resulting aligned MWCNT film inherited anisotropic features from end-to-end connections of the axis direction (the same as the drawing direction) of MWCNTs. The aligned structures were well maintained after immersion for 24 h (rightmost SEM image).



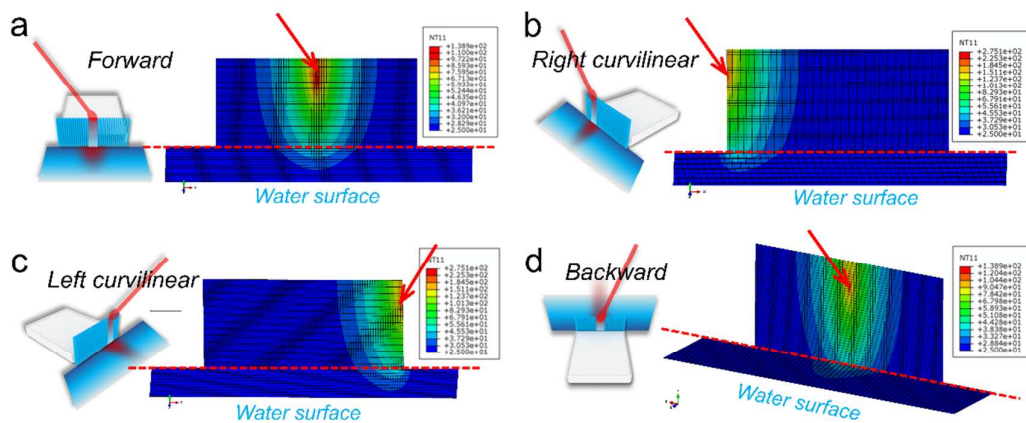
**Figure S2.** Schematic of the propulsion mechanism. The motor navigates forward with the rear part subjected to the illumination. The heated liquid surface (neighboring to the vertically aligned MWCNT film) lowers the local surface tension and retards original  $\gamma_0$  to  $\gamma_1$ . The gradient between  $\gamma_0$  and  $\gamma_1$  generates photo-thermal propulsion, pulling the motor forward.



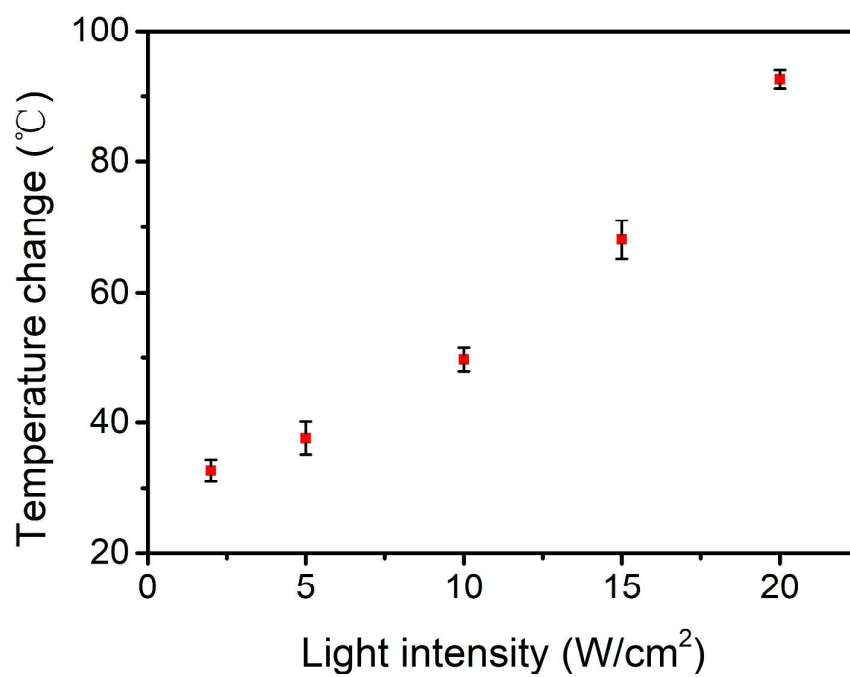
**Figure S3.** Dependence of the mobility on the volume of the motor. An aligned MWCNT film ( $8 \times 6 \text{ mm}^2$ , thickness of  $\sim 800 \text{ nm}$ ) was used to propel the PDMS substrate of motor that consisted of cuboid unit under the irradiation of a light source ( $20 \text{ W/cm}^2$ ).



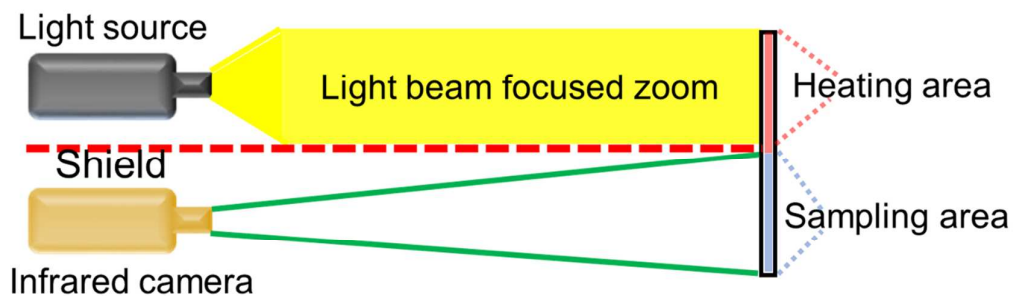
**Figure S4.** Thermal mapping images of a PDMS substrate (area of 8×8 mm<sup>2</sup>) irradiated at the rear part. The substrate was heated but produced no movement.



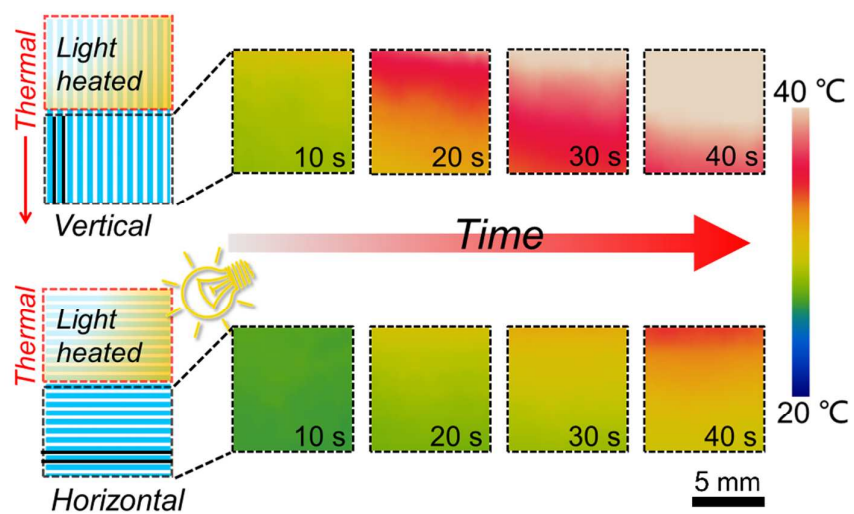
**Figure S5.** Schematic illustrations (left) and finite element simulations (right) on actuations of the light-driving motors irradiated at different sites, producing directional movements toward front, right, left and back from **(a)** to **(d)**, respectively. The simulation results were in line with corresponding motion trails in Figure 2d.



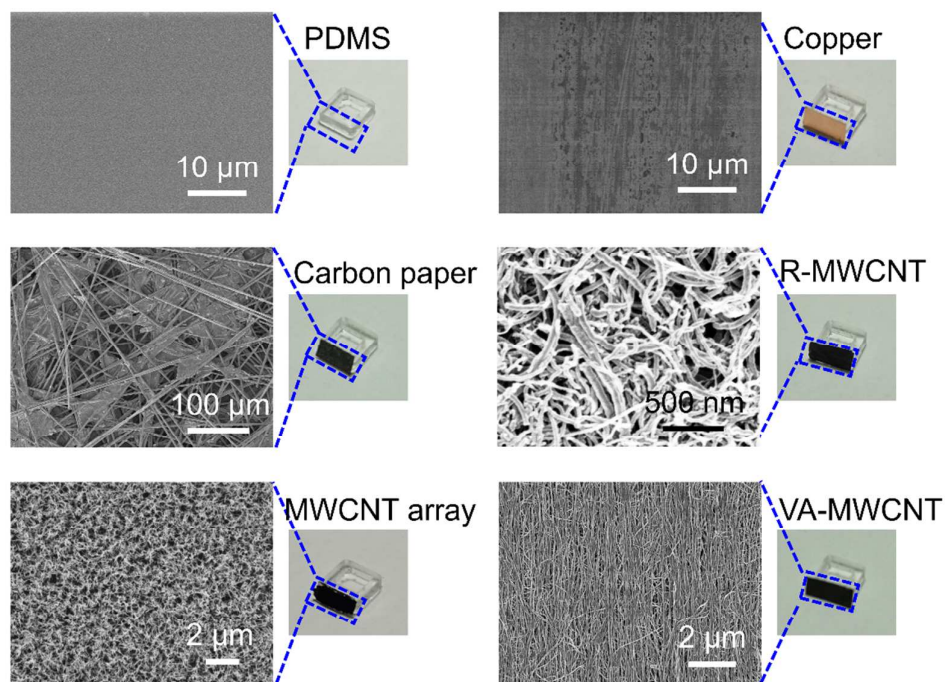
**Figure S6.** Dependence of the temperature change (variation) of the moving light motor on the light intensity of driving light source.



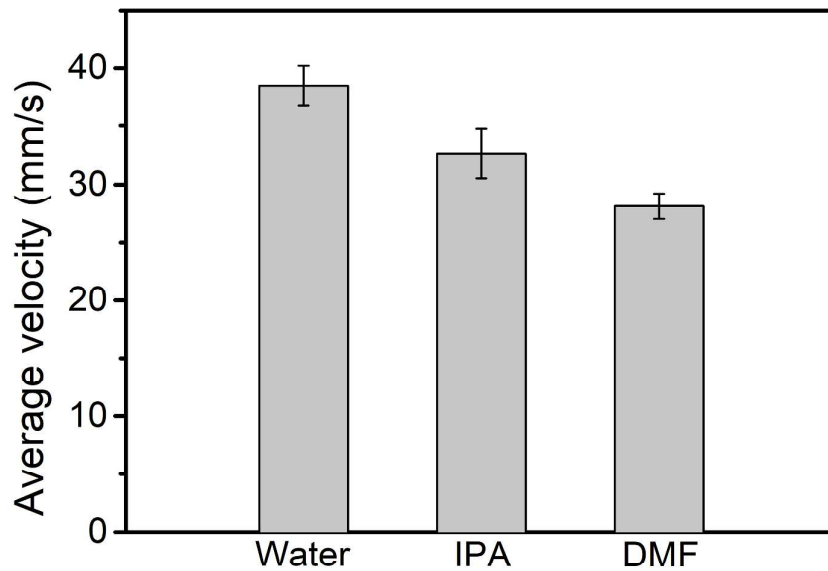
**Figure S7.** Two MWCNT films with different aligned directions were placed side by side at the same height. The top halves of the two films were heated by the same light source, and the bottom halves of them were shielded from heating and continuously monitored by an infrared camera. The captured infrared video of bottom halves depicted the heat diffusion from heated top parts in the vertical direction.



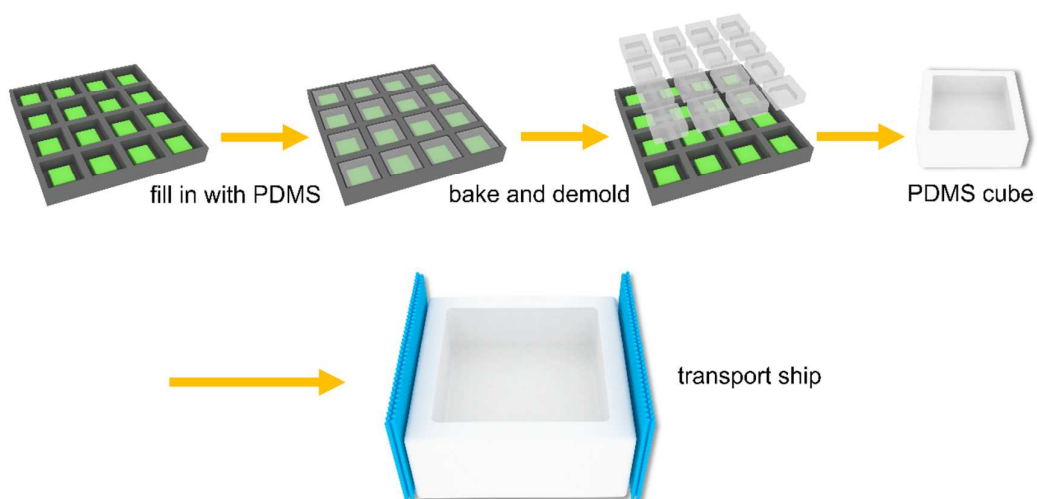
**Figure S8.** Dependence of light-induced thermal transmission on MWCNT-aligned directions (i.e., vertically and horizontally aligned MWCNT film).



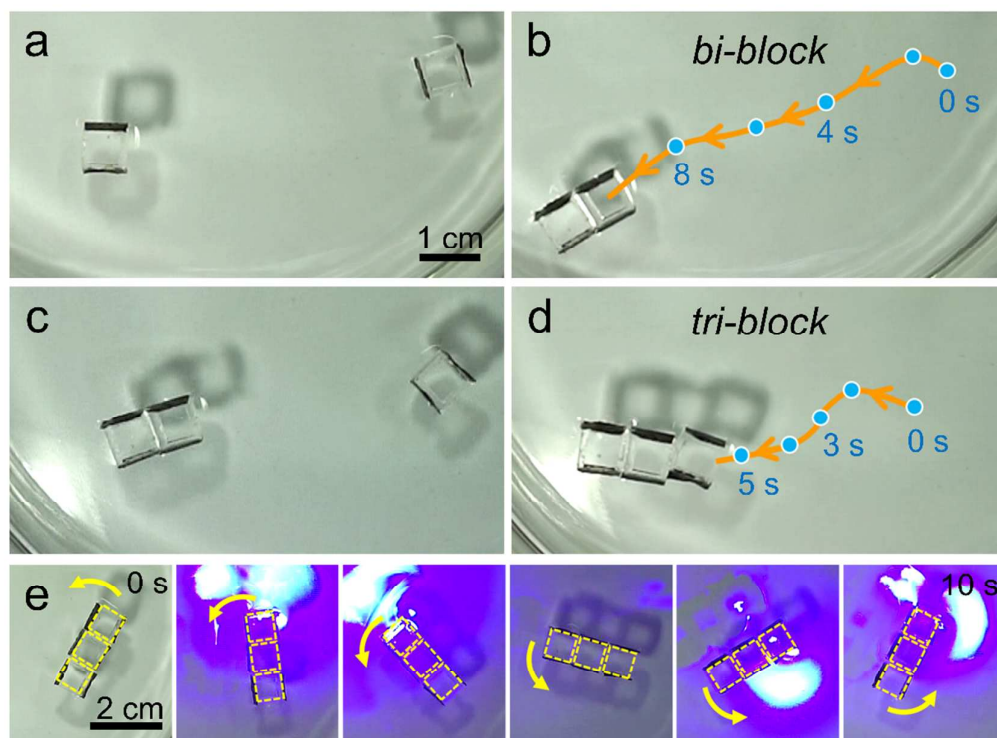
**Figure S9.** Light-driving motors based on six representative materials with blow up SEM images. R-MWCNT: spin-coated randomly dispersed MWCNT film. VA-MWCNT: vertically aligned MWCNT film.



**Figure S10.** Average moving velocities of light-driving motors on different liquid surfaces. Light intensity: 20 W/cm<sup>2</sup>; IPA: isopropyl alcohol; DMF: dimethyl formamide.



**Figure S11.** Schematic illustrating the fabrication process of a light-driving transport ship.



**Figure S12.** Light-manipulated macroscopic assembly. The motors were used as building blocks to form complicated while tunable structures under the guidance of light. **(a)** and **(b)**, The assembly of 2 blocks. **(c)** and **(d)** The assembly of 3 blocks in a line. **(e)** The assembled structure propelled by light irradiation.

## **Captions for Supplementary Movies**

**Movie S1.** Linear motion through the water surface of a light-driving motor when irradiated at the rear MWCNT side.

**Movie S2.** Thermal mapping video of a moving light-driving motor.

**Movie S3.** Simulated thermal distribution of the light-activated motor based on vertically aligned MWCNTs and its neighbouring water surface, with irradiation point at the leftmost side for 1 s (the video playing at a 20% speed).

**Movie S4.** Directional motions manipulated by varying irradiating sites.

**Movie S5.** Light-steered transport boat through an obstacle course.

**Movie S6.** Light-driving macroscopic assembly.

**Movie S7.** Light-driving rotor.