Supporting Information

Near Infrared-modulated Propulsion of Catalytic Janus Polymer Multilayer Capsule Motors

Yingjie Wu, Tieyan Si, Xiankun Lin*, and Qiang He*

Key Laboratory of Microsystems and Microstructures Manufacturing, Ministry of Education,

Micro/Nanotechnology Research Centre, Harbin Institute of Technology, Harbin 150080, China

*E-mail: xiankunlin@hit.edu.cn; qianghe@hit.edu.cn

SI Video 1. The collective video showing autonomous motion of Janus motors in 1%, 2%, 3%, and 5% H₂O₂ solution.

SI Video 2. The start-up of a Janus motor under NIR laser irradiation in 0.1% H₂O₂.

SI Video 3. The start-up and turn-off of Janus motors controlled by NIR laser radiation in 0.05% H₂O₂. The "on/off" cycles were repeated for several times

SI Video 4. The control experiment of the start-up of a Janus motor under NIR laser irradiation without H_2O_2 .



SI Figure 1. SEM image and EDX mapping analysis of a Janus motor before (A, B, C) and after (D, E, F) removing the template. Scale bar = $5 \mu m$.

Description of SI Figure 1

Scanning electron microscopic (SEM) images in SI Figure 1A, D reveal the successful preparation of Janus (PSS/PAH)₅ capsule motors before and after removal of the silica templates. The diameter of (PSS/PAH)₅ capsule motors is approximately 8 μ m corresponding to the size of the silica template. The brighter parts are ascribed to the deposition of the Pt component (SI Figure 1A), and the surface area of the deposited Pt layers is roughly 40% ~ 50% of the total surface area of the capsule motors. The corresponding energy-dispersive X-ray (EDX) mapping analysis (SI Figure 1B, C, E, and F) displays the asymmetric distribution of Pt layers on Janus (PSS/PAH)₅ capsule motors.



SI Figure 2. Optical microscopic (A), fluorescence (B) and CLSM (C) images of Janus capsule motors. Scale bar = $20 \mu m$.

Description of SI Figure 2

After the (PSS/PAH)₅ shells were labeled with fluorescent isothiocyanate (FITC), both fluorescence microscopic and confocal laser scanning microscopic images showed the green Janus structure, confirming the formation of Janus capsule motors and the stability of the assembled PSS/PAH multilayers.



SI Figure 3. (A) The diagram of the NIR beam path of the used microscope. (B) Photograph of the laser setup.

Description of SI Figure 3

The laser irradiation setup was home made. The used microscope was a Olympus BX-53microsocope with a 808nm laser coupled manually into the fluorescence port of the microscope. The used laser was a diode laser (DL-808-200-T from FS-Optics, Shanghai, China) equipped to a external power control unit (APS 30035, ATTEN Instruments, Shenzhen, China). The used Galileian beam expander had one controllable and a fixed lens to vary the beam spot size of the microscope. The spot size used in the microscope was about $1 \times 10 \ \mu m^2$. A simplified scheme of the objective can be observed in SI Figure 4A with a photographic image being shown in SI Figure 4B. The internal microscope optics, as well as the protective filter behind the dichroic is omitted for presentation reasons.



SI Figure 4. Gas chromatography-mass spectrometry (GC-MS) analyses of the component of the bubbles produced by the Janus capsule motors under the NIR illumination.

Description of SI Figure 4

The Janus capsule motors are propelled due to the catalytic decomposition of hydrogen peroxide. The component of the bubbles produced by the Janus capsule motors under the NIR illumination were monitored by GC-MS. The results indicate that the generated bubbles consist of oxygen, and no other gas components were detected.



SI Figure 5. The control experiment of the start-up of a Janus motor under NIR laser irradiation without H_2O_2 . Scale bar = 50 μ m.

Description of SI Figure 5

When dispersing the Janus capsule motors in the pure water and irradiating the motor with the same NIR laser, the Janus motors keep almost motionless. This result indicates that the main driving force for the motor motion is the bubble release due to the catalytic decomposition of the fuel, not the scattering and radiation pressure of the NIR laser or thermophoresis.

Materials

The silica spheres with the diameter of 8 µm were obtained from Microparticles GmbH, Berlin, Germany. Poly (styrenesulfonate) sodium salt (PSS, $M_w = 70,000$), poly-(allylamine hydrochloride) (PAH, $M_w = 70\,000$), fluorescein isothiocyanate (FITC) were purchased from Sigma-Aldrich. Sodium chloride (NaCl), hydrofluoric acid (HF), and hydrogen peroxide (H₂O₂) were used without further purification. FITC-modified PAH (FITC-PAH) was prepared through labeling PAH with FITC according to the literature. The water used in all experiments was prepared in a Milli-Q purification system with the resistivity higher than 18.2 MΩ cm⁻¹.

Preparation of the metal half-shell coated LbL-assembled capsules:

(PSS/PAH)₅-coated particles were first synthesized by the layer-by-layer assembly of polyelectrolyte layers on the surface of SiO₂ particles with a diameter of 8 μm. The SiO₂ particles were alternatively suspended in 2 mg/mL PSS/PAH solution containing 0.5 M NaCl for 15 min under continuous shaking, followed by three repeated centrifugation/washing steps. The (PSS/PAH)₅-coated silica particles were obtained by repeating the above deposition procedure and the outer layer was PAH. To fabricate the Janus structures, we first drop casted a 3 mL colloidal suspension of polyelectrolyte modified silica particles (diluted in ethanol with a 1:5 ratio) onto clean 2 cm² silicon substrates tilted at an angle of ca. 9°. The monolayer is dried via slow evaporation of the solvent at room temperature, allowing the beads to spread into a self-assembled monolayer. The particle-coated substrates were loaded into a vacuum chamber and a 2-nm Cr adhesion layer and 5-nm Pt layer were deposited by electronbeam evaporation (EMITECH K575X) in a vacuum environment at a vapor incidence angle of 30°, resulting in roughly half-coated beads. The silica cores were then dissolved by treatment with 1 M HF (Caution: HF is extremely toxic and can penetrate the skin!). The capsules were purified by three centrifugation/water washing steps. All obtained capsule solutions were stored at 4 °C. Through releasing the capsules into the hydrogen peroxide fuel, micromotors performed self-propulsion by

accumulated O_2 gas. The movement of mivromotor was recorded by microscope coupled with photometric camera.

Theoretical Modeling.

The Janus motor is considered as a standard round sphere. For a halfsphere of Pt film, we cut the asymmetric spherome into many layers of short elementary circular crosssection and performed integration over the height to solve the heat transfer equation in MatLab. The maximal temperature is around the center of the semicircle top, and the temperature of the wide base is lower than the top. In the vicinity of the micromotor, the temperature profile is highly asymmetric contrast the Pt film side and polyelectrolyte film side. Perpendicular to the hemisphere interface, the profile of the temperature becomes symmetric of the center axis of the micromotor. The highly localized temperature is consistent with other theoretical works.^[3-5]

GC-MS analysis

The gas analysis was performed using a GC–MS system Gc6890-Ms5973 (Aglient Technologies, America). The injector temperature was maintained at 280 °C. Splitless injection was employed and the gas injection volume was 1 mL. The oven temperature was set at 38 °C for 2 min, then increased to 100 °C at 3 °C·min⁻¹ and held for 5 min, and then heated at 30 °C·min⁻¹ to 240 °C. Carrier gas was helium (purity > 99.999%) and the flow rate was set at 1.0 mL·min⁻¹. The electron impact (EI) ion source, quadrupole mass analyzer, and the interface temperature were maintained at 280 °C, respectively. Electron impact ionization (70 eV) was utilized. The mass spectra of target compounds were acquired and quantified in the selected ion monitoring (SIM) mode. For the result, the confirmed peak was at the position of relative molecular weight 32, so we affirmed the test gas was oxygen.

Characterization

Scanning electron microscopy (SEM) and energy-dispersive X-ray (EDX) analyses were performed using a Hitachi S-5200 microscope. Copper grids sputtered with carbon films were used to support the sample. Fluorescence images were obtained using a Leica TCS SP5 II confocal laser scanning microscope (CLSM). The excitation wavelength was 488 nm for excitation of FITC dye. The Olympus BX53 fluorescence microscope coupled with a $40\times$ objective and relative software was employed to record the motion of capsule motors. The trajectories of metal half-shell coated capsule motors were analyzed by *Image J* software.

References

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