

A Universal Way to Manipulate Droplet via Light-Fueled Thermocapillary Convection

Hyesun Hwang, Yinggang Zhao, Kun Li, Jaeseung Lee, Cunjing Lv, and Sanghyuk Wooh*

Droplet manipulation plays an important role in scientific research and applications, such as microfluidics and microreactors. Among various driving forces, light-induced thermocapillary convection is a good candidate for propelling liquid droplets. Until now, photothermal additives that convert light into heat have been used as an essential component for droplet manipulation. However, additive-based droplet manipulation relies on inefficient heat transfer and can be applied to limited surfaces and/or droplets. Herein, a new way to manipulate droplets is introduced, directly powered by mid-infrared (MIR) light irradiation. Liquids, such as water and alcohol, exhibit high absorption coefficients for MIR light, efficiently converting it into heat. By using this phenomenon, the MIR laser is applied to realize thermocapillary convection. Liquid molecules at the outer surface of the droplet can spontaneously generate heat fueled by MIR laser, so that powerful thermocapillary convection can be generated within milliseconds. This simple yet effective thermocapillary convection enables highly efficient, controllable, and precise droplet manipulations. No additional additives and/or complicated substrates are required, allowing the method to be versatile for most kinds of liquid droplet and liquid repellent surfaces. Additionally, it is very advantageous for microreactor applications because it does not require a purification process to remove additives.

toward broader applications, such as diagnostics,^[1] bioengineering,^[2,3] and fine chemical synthesis.^[4,5] Droplet motion occurs when the applied driving force exceeds the frictional force. Therefore, surfaces coated with lubricant films have been typically used as substrates to demonstrate droplet manipulation due to the extremely low friction between the droplet and substrate.^[6,7] Additionally, various external stimuli have been explored to induce droplet motion, including acoustic waves,^[8–10] wettability gradients,^[11–13] electric fields,^[14–16] magnetic fields,^[17–19] temperature,^[20–23] and light.^[24–30] Light, in particular, offers contactless, non-invasive, multifunctionality by combining diverse mechanisms, such as photochromic wettability switching,^[24,26] photovoltaics,^[31,32] photopyroelectric effects,^[33–36] and photothermal Marangoni flow.^[37–42] Among them, light-induced thermocapillary convection has recently gained attention due to its rapid response and excellent controllability via localized heating. Thermocapillary convection is a Marangoni flow driven by a temperature

gradient, which moves droplets from a higher to a lower temperature region.

Until now, the manipulation of droplets via light-induced thermocapillary convection has required photothermal materials that absorb light and convert it into heat. These materials have been incorporated into either i) the substrate or ii) the liquid droplet, as a dispersion or as a surface coating materials.^[43–49] Although all methods enable precise control, inefficient propulsion remains a challenge to be overcome. For instance, a droplet on a substrate containing photothermal particles can be locally heated by near-infrared (NIR) laser irradiation.^[28,43–45] However, this approach is inefficient for droplet heating because heat originates from the substrate and transfers to the droplet. In addition, these substrates are complex to prepare, limiting their applicability to versatile droplet manipulation. On the other hand, incorporating photothermal particles into the droplet allows for more direct heat transfer. Dispersion droplets enable droplet motion on conventional lubricant-impregnated surfaces without additional substrate modification.^[46,47,50] However, heat diffusion from the dispersion is still not an optimal mechanism. In microreactor applications, photothermal particles inside the droplet can interfere with the reaction and are considered as impurities that must be

1. Introduction

Method for remotely transporting liquid droplets has emerged as an essential technique for advancing microdroplet research

H. Hwang, J. Lee, S. Wooh
Department of Chemical Engineering
Chung-Ang University
84 Heukseok-ro, Dongjak-gu, Seoul 06974, Republic of Korea
E-mail: woohsh@cau.ac.kr

Y. Zhao, K. Li, C. Lv
Department of Engineering Mechanics
AML
Tsinghua University
Beijing 100084, China

The ORCID identification number(s) for the author(s) of this article can be found under <https://doi.org/10.1002/adfm.202507795>

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removed after the reaction. Alternatively, when photothermal particles are placed on the droplet surface, such as liquid marbles or particle-stabilized emulsions, heat is generated closer to the liquid-air interface, improving thermal efficiency and preserving the inner purity of the droplet.^[48,49,51,52] Nevertheless, such systems still require additives and often involve specific fabrication processes.

Due to the growing interest in clean and material-independent actuation methods, droplet manipulation based on different physical mechanisms, including triboelectric effects and ultrasonic fields, has already been demonstrated without the use of additives.^[53–55] Moreover, although a few attempts have been made to achieve droplet manipulation without additives under light irradiation, the approach has relied on extremely high-intensity irradiation (e.g., 17 W) due to the low light absorption of commonly used substrate or liquid in the NIR region, which may limit their practicality and applicability.^[56] Therefore, to enable a more efficient and generally applicable light-driven approach, it is essential to generate heat directly from the molecules at the surface of the liquid droplet. Herein, we propose a novel and universal strategy to control droplet motion through direct local surface heating of the liquid droplet under mid-infrared (MIR) light irradiation. It is well known that MIR light, in the wavelength range of 2.5–20 μm , is strongly absorbed by various liquids, leading to efficient heat conversion. For example, water exhibits a high absorption coefficient in the MIR region.^[57,58] This absorption characteristic is primarily attributed to the vibrational modes of water molecules, including the bending mode of the H–O–H bond and the stretching modes associated with hydrogen bonding. Specifically, the H–O–H bending vibration and libration mode, related to rotational and vibrational transitions, correspond to wavelengths of ≈ 6.1 μm and 10–20 μm , respectively.^[59–61] Additionally, not only water but also glass and silicone-based materials exhibit photothermal conversion under MIR light due to the Si–O–Si stretching mode.^[62,63]

Under MIR laser irradiation, water molecules at the outer surface of the droplet generate heat, inducing Marangoni flow. This process is highly effective because the temperature gradient is formed directly at the droplet surface. Additionally, the irradiated spot is heated up more than 10 $^{\circ}\text{C}$ within milliseconds, enabling precise and rapid droplet transport. Moreover, MIR laser can also be used to manipulate droplets through indirect heating on a glass substrate. Although droplet manipulation via indirect heating is less efficient than direct droplet heating, it allows for manipulation of multiple droplets and droplet control through backside irradiation and prevents contamination during manipulation. These unique manipulation processes can be applied to pure liquid droplets as well as conventional liquid repellent surfaces without the need for photothermal additives. In this context, the use of the MIR laser irradiation opens up new possibilities for droplet manipulation in a wide range of microreactors and/or microfluidic systems.

2. Results and Discussion

2.1. MIR Light-Fueled Droplet Manipulation

We introduce a method to manipulate droplets by using MIR laser (wavelength: 10.6 μm), which enables droplet surface heat-

ing (Figure 1a). Laser-driven local surface heating induces thermocapillary convection that can propel droplets when it overcomes contact line friction. Since a liquid repellent surface, exhibiting low friction for droplets, facilitates the droplet manipulation, in this study, a lubricated surface (LuS) applied to glass was mainly used as a substrate.^[46,64,65] The LuS contains a uniform silicone oil layer, as a lubricant layer, on the polydimethylsiloxane (PDMS) brush of ≈ 10 nm grafted substrates (Figure 1b). We employed a silicone oil layer of ≈ 3 μm thickness on the glass by spin coating. The top image in Figure 1b indicates a water droplet placed on the LuS glass, characterized by a confocal microscopy. When the droplet contacts the surface, the silicone oil meniscus is immediately formed around the contact line. Due to the mobile silicone oil surrounding the droplet, droplets can easily slide on the LuS with negligible friction. For example, Figure 1c shows a water droplet (5 μL) sliding on the 3 $^{\circ}$ tilted LuS glass. Indeed, the droplet can move even at the tilted surface below 1 $^{\circ}$. For this reason, the LuS was applied as a major substrate to manipulate droplets.

To induce local surface heating, the MIR laser was applied. Due to its high absorption efficiency by the H–O–H bending mode, water quickly generate heat under MIR laser irradiation. In addition, glass and silicone produce heat, attributing to the Si–O–Si stretching mode. The absorption coefficient at 10.6 μm of water and glass is ≈ 800 and 10^3 cm^{-1} , corresponding to the absorption depth of 12.5 and 10 μm , respectively.^[62,66] This demonstrates that most of the laser energy is absorbed within a thin surface layer. The photothermal effect under MIR laser irradiation was examined by using an infrared (IR) camera, which identified a clear temperature increase at the surfaces of water and the LuS glass (Figure 1d). The temperature of the irradiated spots increased immediately and was saturated almost within 5–10 s. Figure 1e shows the temperatures of heated spots on substrates and/or liquids, observed after 40 s MIR laser irradiation from 20 $^{\circ}\text{C}$. Details with time-dependent temperature changes are provided in Figure S1a–e (Supporting Information). Water and the glass-based surfaces absorb MIR light and exhibit highly effective heat generation. Note that the temperature profiles of water and silicone oil were determined by measuring the temperature of the liquid in a small petri dish (liquid volume: 200 μL), as shown in Figure S1f (Supporting Information). Due to convection, the measured temperature may be lower than the actual temperature. Meanwhile, heat production by silicone oil is not as strong as other tested materials. This is because the Si–O–Si bonds in silicone oil do not resonate with the 10.6 μm wavelength, resulting in weak absorption.^[67] The stretching modes of the Si–O–Si bonds in silicone material primarily correspond to wavelengths in 9.1–10 μm range. However, as silicone oil consists of flexible Si–O–Si chains bonded with CH_3 groups, it exhibits high flexibility due to the free rotation of its randomly oriented Si–O bonds. These intermolecular interactions contribute to their low absorption characteristics under MIR irradiation. In the case of the LuS glass, temperature variation is almost the same as the bare glass. We expect that the heat on the LuS glass is mainly generated by glass under the thin silicone oil layer, ≈ 3 μm . The heating as well as cooling can occur within a few seconds, as shown in Figure 1f. The increased temperature on water and the LuS glass immediately cooled down to their initial temperatures after turning off the laser. In addition, we verified the heating/cooling can

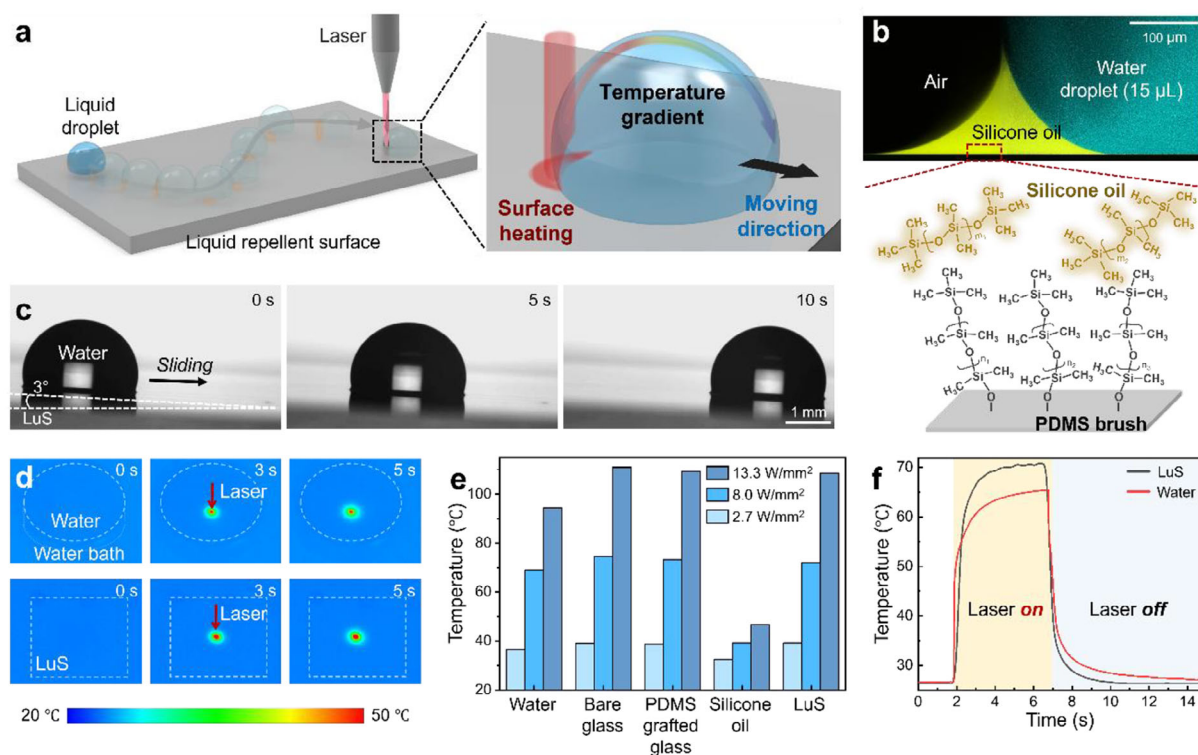


Figure 1. a) Schematic illustration of MIR-fueled droplet manipulation on a liquid repellent surface. The laser-induced temperature gradient is formed on the surface of the droplet or the substrate. The liquid droplet moves from the irradiated region to the unirradiated region. b) Water droplet on the LuS glass characterized by a confocal microscopy (top) and structure of the LuS composed of a PDMS brush and silicone oil layer (bottom). c) Images of the sliding water droplet (5 μL) on the LuS glass. The tilting angle is 3° . d) Time-sequence IR images of water in a small petri dish (top) and the LuS (bottom) under the laser intensity of 8.0 W mm^{-2} , respectively. e) Maximum temperature of the heated spots on liquids and/or substrates observed after 40 s under different irradiation intensities (2.7, 8.0, and 13.3 W mm^{-2}). Water and silicone oil were measured using a small petri dish with a liquid volume of $200 \mu\text{L}$. f) Heating and cooling curves of water (red line) and the LuS glass (gray line) under the laser intensity of 8.0 W mm^{-2} for 5 s.

be repeated over 10 cycles (Figure S2, Supporting Information), which shows the feasibility of precise droplet manipulation in a well-controlled manner by MIR irradiation.

2.2. Powerful Thermocapillary Convection with Rapid Response Time

By using the MIR-fueled local surface heating of water droplets, here, we demonstrated droplet manipulation on the LuS glass (Movie S1, Supporting Information). To control the velocity, the MIR laser was moved by fixing it at an axial motorized stage (Figure S3, Supporting Information). Figure 2a,b shows the sliding motion of water droplets (volume: $15 \mu\text{L}$), propelled by the MIR laser of different intensities, a) 2.7 W mm^{-2} and b) 8.0 W mm^{-2} , respectively. Infrared (IR) thermal camera images (right images of Figure 2a,b) confirmed that only the irradiated spot was heated causing the temperature gradient at the surface of droplet. The temperature difference between the irradiated region and the counter edge of the droplet depended on the laser intensity, which was ≈ 5.2 and 9.0°C by the laser intensity of 2.7 and 8.0 W mm^{-2} , respectively (Figure S4, Supporting Information). From the images, we found that the larger temperature difference induces faster motion. It was investigated with various sizes of droplets under different laser intensities (Figure 2c).

Increasing the laser intensity enhanced thermocapillary convection, thereby increasing the velocity of the droplet. In addition, droplets of the smaller volume showed faster velocity under the same irradiation condition (Table 1).

To study the underlying mechanism of droplet motion, by using the above results, we analyzed the forces acting on the droplet. According to previous literature, three distinct forces are exerted on the droplet: Marangoni force (F_M),^[22,45,68] Young's force (F_Y),^[7,69] and hydrodynamic resistance force (F_H).^[7,56] The Marangoni force (F_M) is defined as^[22,45,68]

$$F_M \approx \pi \left(\frac{D}{2} \right)^2 \left(\frac{dT}{dx} \right) \left(\frac{d\gamma_{la}}{dT} \right) \quad (1)$$

where D is the contact diameter of the droplet, dT/dx is the temperature gradient along the x -axis, and $d\gamma_{la}/dT$ is the surface tension gradient at the liquid-air interface by temperature. To calculate the temperature gradient along the x -axis, the temperature difference at the receding and advancing points ($\Delta T = T_A$ (temperature at the receding point (A)) $- T_B$ (temperature at the advancing point (B)) under the 8.0 W mm^{-2} intensity of laser irradiation was measured (Figure 2d). The temperature differences were ≈ 7.9 , 9.0 , and 10.8°C for 5, 15, and $30 \mu\text{L}$ droplets, respectively. Since Marangoni flow is driven by surface tension gradient along the droplet surface, the temperature gradient was evaluated

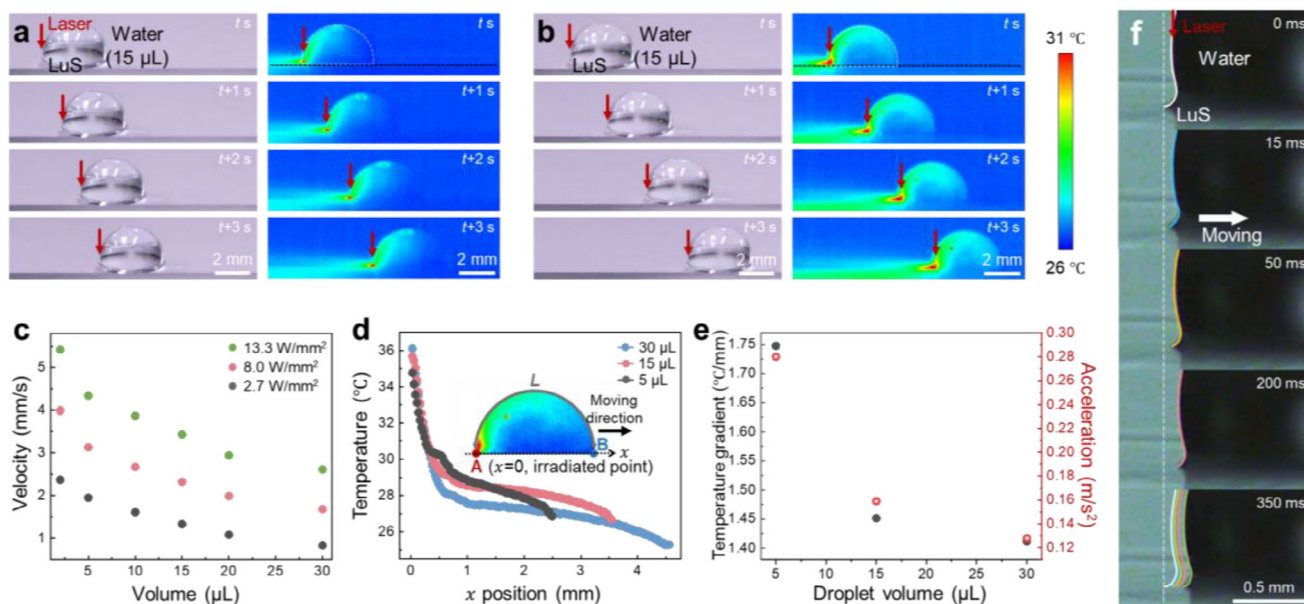


Figure 2. Sliding motion of water droplet on the LuS glass under the laser intensity of a) 2.7 W mm^{-2} and b) 8.0 W mm^{-2} , respectively. The images on the left and right panels in (a) and (b) represent the optical and IR images, respectively. c) Velocity variation of water droplets on the LuS glass as a function of volume (2, 5, 10, 15, 20, and 30 μL) and irradiation intensity (2.7, 8.0, and 13.3 W mm^{-2}). d) Temperature profile from the receding (A) to advancing (B) points when droplet was manipulated under the laser intensity of 8.0 W mm^{-2} along the x -axis as a function of volume. e) Variation of the temperature gradient ($\Delta T/L = (T_A - T_B)/L$) and the acceleration ($a = \text{Marangoni force } (F_M)/\text{respective mass}$) with droplet volume based on (d). f) Pictures show a fast response of droplet on the LuS glass under the laser intensity of 8.0 W mm^{-2} . The colored lines indicate the shape changes at the rear side of droplet with time.

along the droplet contour rather than the horizontal x -axis direction, in order to more accurately reflect the direction of thermocapillary flow.^[46] Hence, the temperature gradients, calculated by dividing the temperature difference by the contour length (L), for each droplet were 1.75 , 1.49 , and 1.41 K mm^{-1} . The surface tension gradient of water by temperature, characterized by surface tensions at various temperatures, was $-0.167 \text{ mN mK}^{-1}$ (Figure S5, Supporting Information). F_M could be estimated with the above values. Then, accelerations were assessed by F_M , which were 0.28 , 0.16 , and 0.13 mm s^{-2} for the droplets of 5, 15, and 30 μL , respectively (Figure 2e).

Once droplets moved, a hydrodynamic resistance force (F_H) was generated as resistance, which can be expressed as below^[7,56]

$$F_H \propto \pi (\mu_l + \mu_o) v (D/2) \quad (2)$$

where μ_l is the viscosity of the liquid droplet, μ_o is the viscosity of the oil, and v is the velocity of the droplet. Since F_H is pro-

portional to the droplet diameter, as given by Equation (2), it is greater for the larger droplets than the smaller droplets. Therefore, the smaller droplets exhibited faster motion, as shown in Figure 2c, by the larger acceleration with the smaller resistance. Moreover, the resistance resulting from contact angle hysteresis (F_Y) could also be occurred by the movement due to the asymmetric deformation of the droplet, which is described as^[7,69]

$$F_Y = D (\cos \theta_{adv} - \cos \theta_{rec}) \gamma_{la} \quad (3)$$

where θ_{adv} is the advancing contact angle, θ_{rec} is the receding contact angle, and γ_{la} is surface tension at the liquid-air interface. On the LuS, however, low contact angle hysteresis is close to zero, resulting in a negligibly small F_Y .

In addition to photothermally induced thermocapillary convection, Rayleigh-Bénard convection attributed to density variation can be produced by the heating. The relative contributions of Rayleigh-Bénard convection and thermocapillary convection are described by the Bond number (Bo). The Bond number is

Table 1. Wetting properties of water droplets on the LuS glass. The intensity of the MIR laser for propulsion was 8.0 W mm^{-2} . All measurements were conducted 5 times to obtain average values.

	Water					
Volume [μL]	2	5	10	15	20	30
Contact diameter [mm]	1.8 ± 0.01	2.5 ± 0.06	3.0 ± 0.02	3.5 ± 0.03	3.9 ± 0.03	4.6 ± 0.02
Velocity [mm/s]	4.0 ± 0.1	3.1 ± 0.05	2.7 ± 0.07	2.3 ± 0.05	2.0 ± 0.05	1.7 ± 0.08
Droplet height [mm]	1.2 ± 0.01	1.5 ± 0.03	1.8 ± 0.02	2.1 ± 0.01	2.2 ± 0.01	2.4 ± 0.01
Contour length [mm]	3.3 ± 0.02	4.5 ± 0.03	5.5 ± 0.01	6.2 ± 0.01	6.8 ± 0.02	7.7 ± 0.03

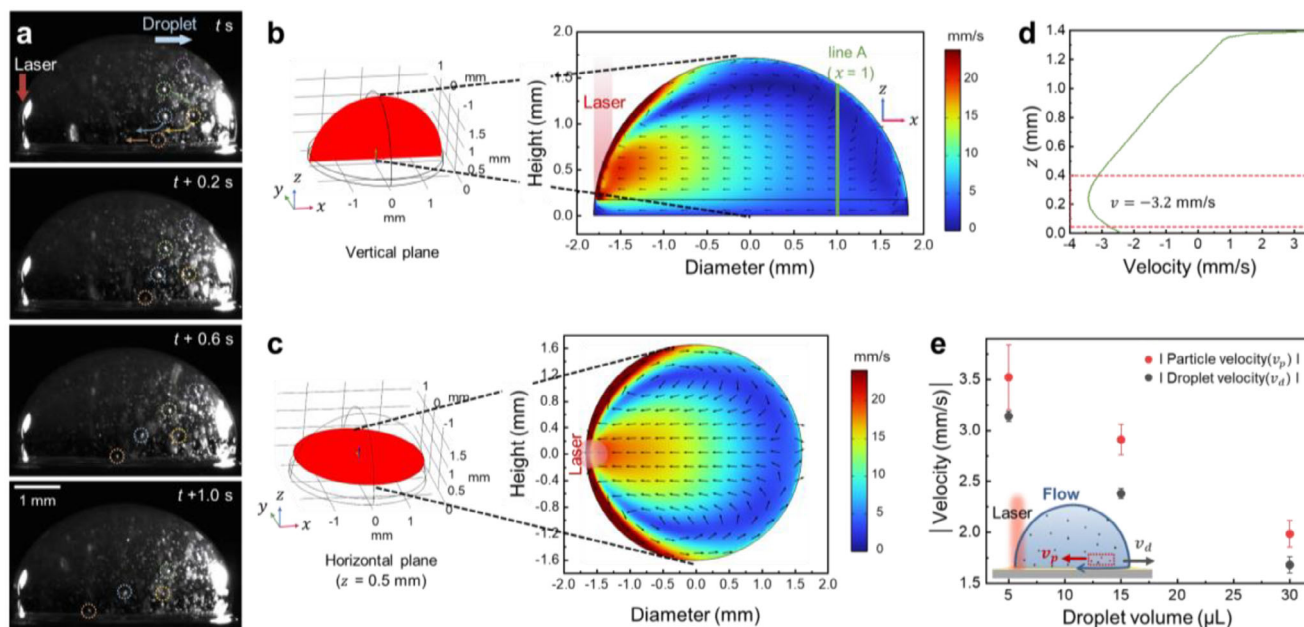


Figure 3. Internal flow of the water droplet induced by the laser on the LuS glass. a) Lateral image of movement of tracer particles (polystyrene particles, 1–50 μm) inside a moving droplet (volume: 30 μL) on the LuS glass under the laser intensity of 8.0 W/mm^2 . Flow simulation at b) the vertical (x - z) plane of the droplet and c) horizontal (x - y) plane where $z = 0.5$ mm, respectively. d) Velocity fields at the vertical (x - z) plane and corresponding velocity variations along z -axis at line A ($x = 1$, green). e) Comparison of the relative velocity of the water droplet motion and tracer particles near the bottom of the droplet. The inset scheme presents the internal flow of the tracer particles in side-view. v_d and v_p indicate velocity of droplet and tracer particles near the bottom of the droplet, respectively.

proportional to the ratio of the buoyancy force to the surface tension force.^[70–72]

$$Bo = \frac{\beta g \rho l^2}{|\mathrm{d}\gamma/\mathrm{d}T|} \quad (4)$$

where β is the thermal expansion of the liquid, g is the gravitational acceleration, ρ is the density of the liquid, and l is the characteristic length. The characteristic length l is defined as $l = \frac{V_d}{\pi(D/2)^2}$, where V_d is the volume of the droplet. When Bo is less than 1, the thermocapillary convection is dominant. Conversely, when Bo exceeds 1, the effect of gravity gradually becomes more significant. Bo , here, a water droplet of 30 μL , which is the largest droplet size, is less than 0.08. Therefore, in this study, we expect that F_M is a major driving force in manipulating droplets.

Meanwhile, MIR-fueled strategy demonstrates 2–5 times faster motion compared to previous Marangoni flow strategies for droplet manipulation, as depicted in Figure 2c and Figure S6 (Supporting Information). This outstanding performance is due to the direct generation of heat by molecules at the outermost layer of the droplet. In general, light-induced methods achieves relatively faster droplet motion than other Marangoni flow-driven methods. Figure S7a (Supporting Information) illustrates the early-stage of light-driven method using photothermal materials incorporated into substrates.^[40,43,45] Upon light irradiation, photothermal materials in substrate first absorb the light and convert it into heat, which is then transferred to the overlying droplet, leading to motion. Although this method could be applied to various liquid droplets, the indirect heating mechanism of droplet through the substrate resulted in inefficient thermo-

capillary convection due to light and slow response. Recently, the method using photothermal dispersion demonstrated more efficient motion by generating heat from the photothermal particles inside the droplet (Figure S7b, Supporting Information).^[46,47,50] This method allows manipulation of droplets on ordinary liquid repellent surfaces without the need for additional photothermal functions inside the substrate. In this study, molecules at the top surface of the droplet directly absorb light and convert into heat (Figure S7c, Supporting Information). This direct surface heat generation enables highly efficient light-induced thermocapillary convection, leading to rapid droplet motion and immediate light response with no photothermal materials either on the substrate or in the liquids. The rapid light responsivity was verified by a high-speed camera that the droplet started to move within the order of milliseconds after MIR irradiation (Figure 2f; Figure S8, Supporting Information). The rear side of the droplet, which was the irradiated region, receded after ≈ 15 ms MIR irradiation, and the entire droplet moved distinctly after ≈ 50 ms irradiation. This rapid light responsivity, realized by effective surface heat generation, will be the key to enable precise and accurate droplet manipulation.

2.3. Flow by Thermocapillary Convection

The flows by thermocapillary convection were thoroughly studied by using water droplets with two different tracer particles: i) hydrophobic and ii) hydrophilic particles. The vinyl-terminated polydimethylsiloxane grafted TiO_2 particles were used as the hydrophobic tracer particles, designed to spread over the surface of

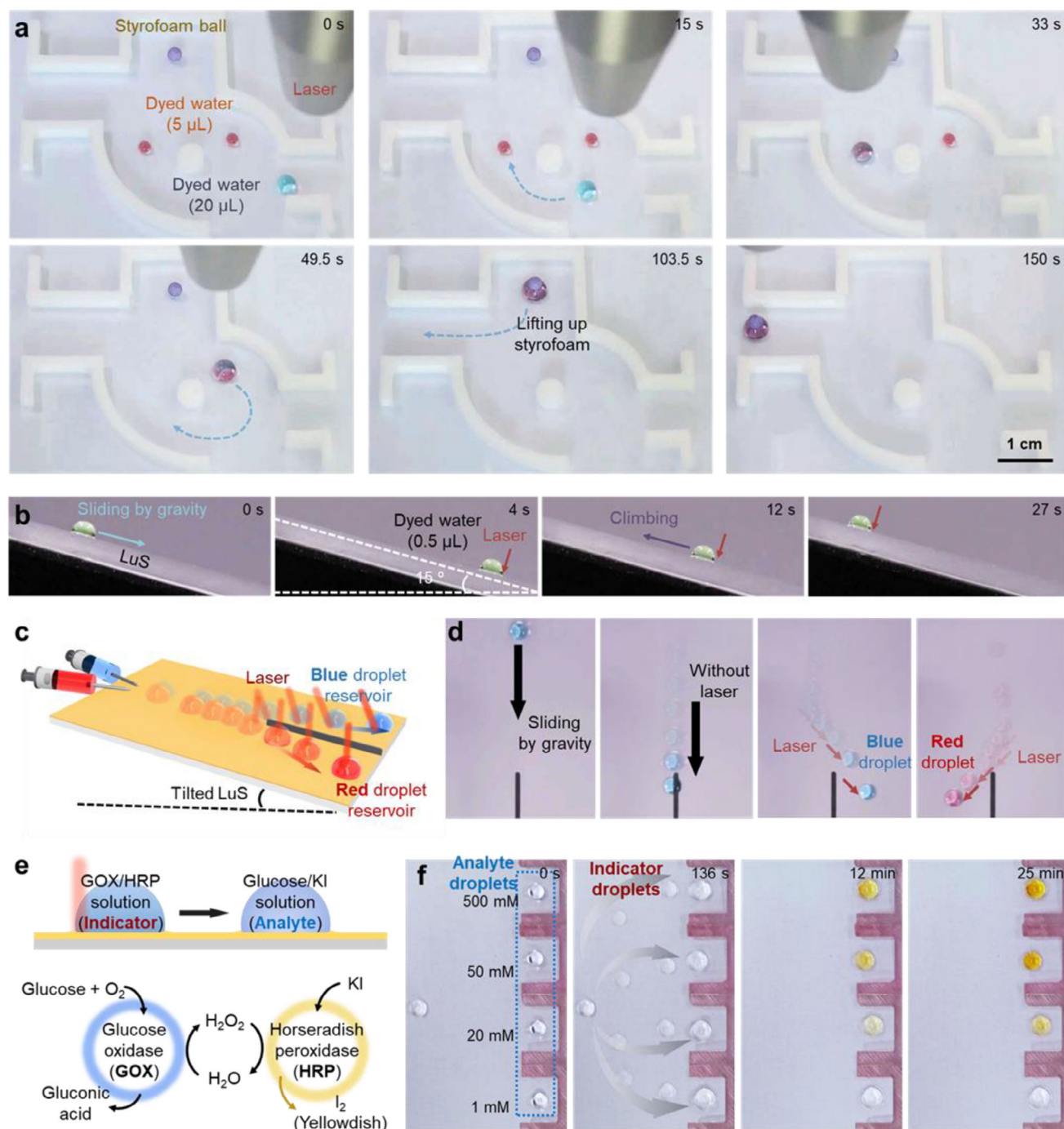


Figure 4. Droplet manipulation for practical applications under laser irradiation (intensity: 8.0 W mm^{-2}). **a**) Complex laser-induced droplet motion, including coalescence and cargo motion. **b**) Anti-gravity motion of the water droplet (volume: $0.5 \mu\text{L}$) on the LuS glass (tilting angle: 15°). **c**) Schematic showing template-free selective sorting of microdroplets into the target reservoirs. **d**) Sliding water droplets (volume: $10 \mu\text{L}$) are sorted on the tilted LuS glass (incline angle: 2°) by the laser. **e**) Scheme illustration of the GOX/HRP cascade reaction by laser-induced droplet manipulation (top). Glucose/KI solutions and GOX/HRP are used as analyte and indicator, respectively. Enzymatic reactions and the colorimetric method for glucose detection using KI as a chromogen (bottom). **f**) Glucose assay depending on the glucose concentration (1, 20, 50, and 500 mM) with time. The yellowish color represents the progress of cascade reactions.

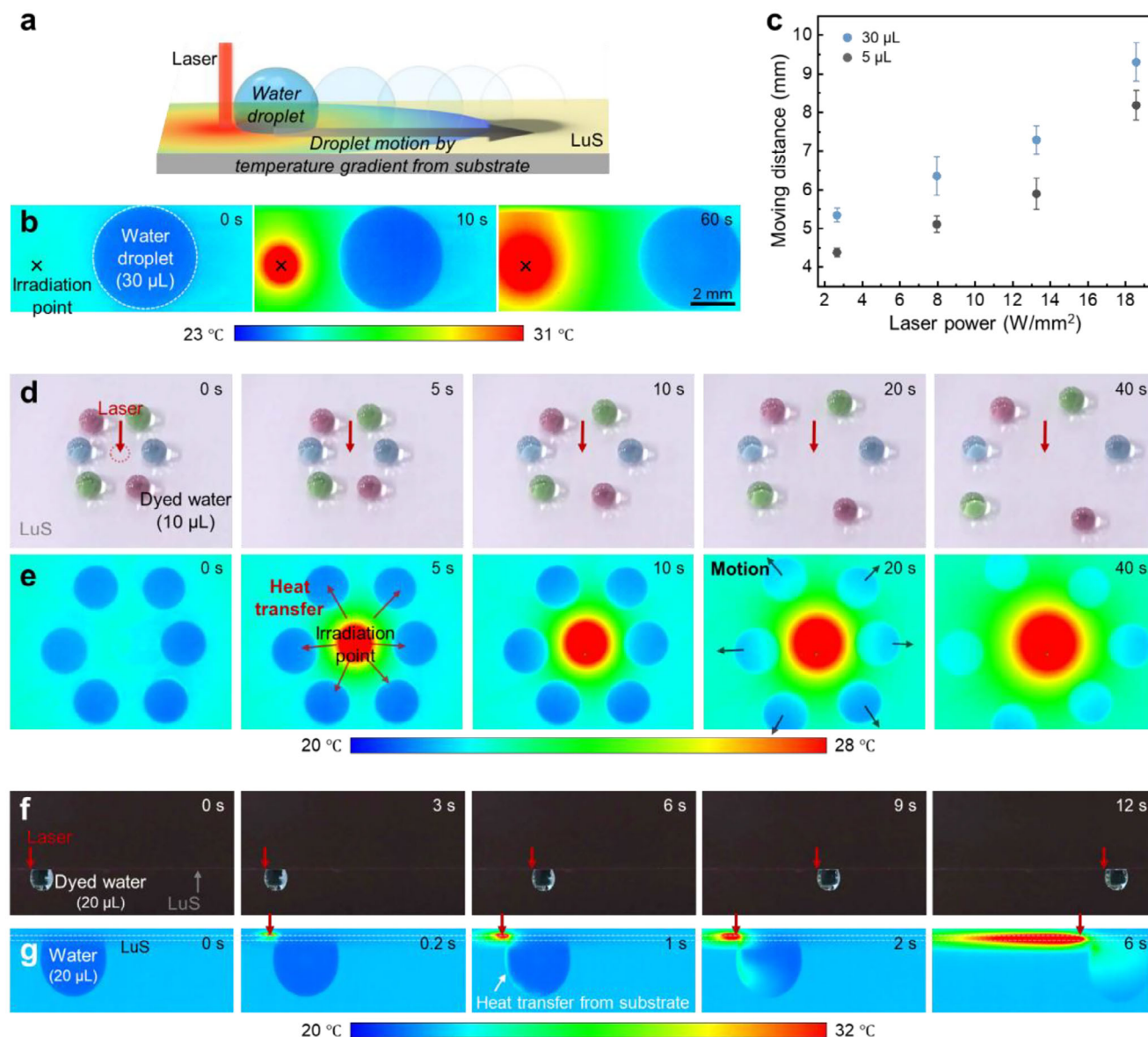


Figure 5. a) The illustration shows droplet manipulation by indirectly heating the substrate under MIR laser irradiation. b) IR images of droplet manipulation by the temperature gradient induced from the substrate. c) Maximum moving distance of water droplets (volume: 5 and 30 μL) on the LuS glass as a function of the laser intensity (2.7, 8.0, 13.3, and 18.6 W mm^{-2}) for 1 min of laser irradiation time. d) Time-sequence optical images of the motion of six water droplets (10 μL) under the single-focused laser irradiation positioned at the center of the droplets. e) IR images of (d). f) Transport of the hanging water droplet (volume: 20 μL) on the inverted LuS glass by the laser. The laser irradiated the top surface of glass. g) IR images of (f).

the droplet, showing directions and velocities of the surface flows of the droplet under MIR laser irradiation (Movie S3, Supporting Information). In contrast, the internal flows were monitored using the hydrophilic tracer particles (1–50 μm , polystyrene particles), which remain within the bulk of the droplet (Figure 3a; Movie S3, Supporting Information). The tracer particles clearly demonstrated the circular flows by thermocapillary convection that started from the MIR laser-irradiated region. The internal flow near the bottom of the droplet appeared in the opposite direction to the surface flow.

In addition, we conducted the flow simulation to estimate the speed and internal flow by thermocapillary convection. The sim-

ulation was performed by solving the Navier–Stokes equations and the energy equation of the flow field with COMSOL Multiphysics (COMSOL 6.1) for a 15 μL water droplet (diameter: 3.5 mm, height: 2.5 mm) under the laser intensity of 8.0 W mm^{-2} (Figure 3b,c).^[73,74] Within the model, the flow was simulated by photothermally induced thermocapillary convection, based on experimentally obtained temperature values. Within this model, the MIR laser irradiation was considered as a heat source, initiating the flow. The flow field satisfied the Marangoni boundary conditions at the liquid–air interface, and the movement of droplet was also considered. For more details on the simulation, see Figure S13 and Note S4 (Supporting Information). The black

arrows in the simulated velocity fields indicate the velocity vectors; the magnitude is represented by color. The velocity vector fields were presented in the vertical (x - z) plane (Figure 3b) and the horizontal (x - y) plane where $z = 0.5$ mm (Figure 3c). The flows in both planes display a similar tendency as the experimental flow tracking results, providing additional evidence of the internal flow of the droplet by photothermally induced thermocapillary convection (Figure S14; Movies S3 and S4, Supporting Information). Moreover, the velocity profile could be obtained from the simulation. The velocity variation along the z -axis, from $z = 0$ to $z = 1.4$ mm, at the vertical plane where $x = 1$ mm (indicated as line A in Figure 3b) was shown in Figure 3d. Positive values present the flow velocities in the x -direction from the rear side (MIR irradiated region) to the front side of the droplet, while negative values indicate the opposite direction. We found that these simulated flow velocities matched well with the experimental results with a difference of only 10–15%. For example, the velocity of the tracer particles (v_p) near the bottom of the droplet (volume: 15 μ L) within the area marked by the red box in Figure 3d,e (ranging from $z = 0.05$ to $z = 0.4$ mm, where $x = 1 \pm 0.3$ mm) was -2.9 ± 0.2 mm s $^{-1}$, while the estimated flow velocity by the simulation at the same region was ≈ -3.2 mm s $^{-1}$. This reliable flow simulation helps to predict the droplet movement because the droplet moving speed is attributed to the speed of thermocapillary convection. By using the flow tracking results, we evaluated the absolute values of v_p and the droplet velocity (v_d) of droplets of three different volumes (Figure 3e). Both $|v_p|$ and $|v_d|$ were smaller with larger droplets. The decreasing tendencies of them were nearly the same, which verified the correlation of flow and droplet speeds. Obviously, $|v_d|$ exhibited slightly lower values than $|v_p|$ for all droplets due to slip at the liquid-substrate interface and friction.

2.4. MIR Laser-Fueled Precise Droplet Control

The effective thermocapillary convection by the MIR laser irradiation allows precise droplet motion control. Droplets can move and change direction immediately by controlling the position of the laser due to the fast photothermal response. It was demonstrated with a droplet guided by the laser along an arbitrary trajectory in a complex structure (Figure 4a; Movie S5, Supporting Information). Freely movable droplet showed merging with other droplets and lifting solid matter, a styrofoam ball. The motion by strong thermocapillary convection demonstrated droplet climbing up a 15° tilted slope. On the tilted LuS glass, the droplet initially slid down due to gravity. When the MIR laser irradiation was focused on the center of the rear side of the sliding droplet, the droplet slowed down and then started to move upward. The laser beam continuously followed the droplet behind, maintaining its upward motion by overcoming gravity (Figure 4b; Movie S5, Supporting Information).

Moreover, we designed a template-free droplet sorting system that remotely directs droplets to their target sites. Water droplets placed on the inclined LuS glass slid down straight. In this scenario, gravitational force is the sole driving force for droplet movement. The thermocapillary convection applied perpendicularly to the gravitational force alters the sliding direction. Sliding in different diagonal directions can be easily induced by ir-

radiating the MIR laser to the side of the droplet. As shown in Figure 4c,d and Movie S6 (Supporting Information), we demonstrated the separation of red and blue droplets sliding down the inclined LuS glass (incline angle: 2°) into the reservoirs on the left and the right, respectively. The laser was irradiated on either the left or right side of the sliding droplet in order to guide it toward the desired target reservoir. This flexible directional control capability suggests a novel strategy for sorting droplets without as-prepared channels or templates. Additionally, it can be applied to droplet microreactors. Figure 4e illustrates a droplet performing an enzymatic cascade reaction involving glucose oxidase (GOX) and horseradish peroxidase (HRP).^[75] This reaction is commonly used in colorimetric assays for glucose detection. In the presence of GOX, glucose is oxidized by oxygen (O_2) to gluconic acid, producing hydrogen peroxide (H_2O_2). Subsequently, potassium iodide (KI) is converted to iodine (I_2) by HRP, which consumes hydrogen peroxide, resulting in a yellowish color. Here, water droplets containing different glucose concentrations (1, 20, 50, and 500 mM) and KI (50 mM) were used as analytes and placed separately on the LuS glass. Then, we guided the indicator droplets toward the analyte droplets containing a mixture of GOX/HRP (5 mg mL $^{-1}$; volume ratio of GOX to HRP is 4:1) using the laser. By merging indicator droplets with analyte droplet, the glucose concentration could be distinguished (Figure 4f; Movie S7, Supporting Information); the observed color intensity changed over time depending on the glucose concentration, with higher analyte concentrations yielding a darker yellowish color, showing successful analytical demonstration. For this microreactor system having enzymes, amount of evaporation and enhanced temperature should be considered. The water droplet exhibited $\approx 6\%$ weight loss under the laser intensity of 8.0 W mm $^{-2}$ for 1 min (total travel distance: ≈ 8 cm) (Figure S9 (Supporting Information)). In addition, the maximum temperature at the rear side of the moving droplet is $\approx 36^\circ\text{C}$, observed by the IR camera (Figure 2d). Because Marangoni flow is immediately generated once the molecules at the liquid surface are few degrees heated, the temperature of irradiation point is saturated in the range of 30–40 °C. We believe this temperature is a biologically safe range, especially those not extremely heat-sensitive.

2.5. Indirect Droplet Heating-Induced Droplet Manipulation

Furthermore, the thermocapillary convection of the droplet was induced by the temperature gradient of the substrate (Figure 5a). MIR light can also heat certain substrates, such as glass and wafer. Using these substrates, we demonstrated droplet manipulation via indirect heating with the MIR laser. Upon laser irradiation of the LuS glass, the generated heat is radially transferred to the droplet through conduction, propelling the droplet, as characterized by the IR camera (Figure 5b). So far, this indirect heating for droplet manipulation has only been demonstrated on surfaces containing photothermal materials.^[40,43,47] The MIR laser enables droplet manipulation on conventional liquid repellent surfaces. The plot in Figure 5c presents the displacement of water droplets as a function of laser intensity. In the experiments, the laser was initially irradiated 3 mm behind the rear side of the droplet. The irradiation position was not changed;

therefore, droplets stopped at certain positions. A similar result was also confirmed on the lubricated wafer, as shown in Figure S15 (Supporting Information). Obviously, stronger laser intensity propelled droplets farther. Larger droplets moved more effectively because of the greater temperature difference between the front and rear sides of the larger droplet.^[21,27] This confirmed that droplet motion via substrate heating was driven by thermocapillary convection of the droplet rather than the Marangoni flow of the lubricant. If droplet motions were caused by lubricant flow, droplets of different sizes would travel the same distance driven by this flow.^[21]

We studied the temperature distribution of water droplets and substrate in detail by using the IR camera (Figure S16, Supporting Information). The temperature at the rear side of the droplet (10 μL) gradually increased, while the temperature at the front side remained constant until 9.5 s after laser irradiation. When the temperature difference between the rear and front sides reached $\approx 1.24^\circ\text{C}$, the droplet began to move and accelerated. Afterward, at a certain point, droplet motion eventually stopped when the temperature gradient was no longer sufficient to drive the droplet. This substrate heating method has the advantage of inducing movement of multiple droplets with a single irradiation. We demonstrated multiple droplet manipulation by positioning the laser at the center of six droplets (Figure S4d; Movie S8, Supporting Information). The generated heat spread in all directions and was transferred to the droplets, inducing radial droplet migration with a single-focused laser irradiation. Moreover, the substrate heating from the backside generated thermocapillary convection in the droplet. This was demonstrated with the droplet suspended upside-down on the LuS glass (Figure 5f; Movie S8, Supporting Information). The laser was irradiated from the top, and the generated heat was conducted through the glass to the bottom, creating the temperature gradient in the hanging droplet (Figure 5g). Due to this successful movement by indirect irradiation, MIR fueled droplet manipulation could be a promising approach for controlling droplets in closed system with covers, such as microfluidic systems.

2.6. Droplet Sliding and Rolling on Various Surfaces

Droplet manipulation using the MIR laser was applied to various liquid repellent surfaces. For example, we described droplet motion on convectional liquid repellent surfaces, such as lubricant impregnated surface (LIS) and superhydrophobic surface.^[76–80] Droplet motion on the LIS, which is similar to the LuS but typically contains a hydrophobic porous structure under the lubricant layer, exhibited a sliding motion similar to that on the LuS (Figure S17a, Supporting Information). The difference was that the moving speed of the droplet on the LIS was slightly slower than that on the LuS because of the roughness of the porous structure under the droplet. A droplet placed on a superamphiphobic surface showed rolling under MIR laser irradiation (Figure 6a). We prepared a fluorinated etched aluminum substrate as the superamphiphobic surface, which exhibits an apparent water contact angle of over 160° (Figure 6b,c). Due to small contact lines and high apparent contact angles, droplets have traditionally been difficult to manipulate on such hydrophobic surfaces. Here, a sufficient temperature gradient to manipulate droplets along the

liquid-air interface was generated by the MIR laser due to its highly effective localized heating. MIR-fueled droplet manipulation on the superamphiphobic surface was characterized by a high-speed camera (Figure 6d). A high intensity (26.6 W mm^{-2}) laser irradiation was employed instantaneously for a few milliseconds ($\approx 5\text{ ms}$). Then, the droplet rolled on the superamphiphobic surface, as confirmed by the movement of the attached hydrophobic glass beads when the droplet passed. Figure 6e and Movie S9 (Supporting Information) also show a rapid rolling motion driven by laser irradiation. When the propulsion force overcame the static friction, the droplet immediately rolled, visualized also by the IR camera. Moreover, we observed a similar droplet motion on another superamphiphobic surface. A droplet placed on the soot-templated superamphiphobic surface, showing a high apparent water contact angle of over 165° , could also be manipulated by rolling (Figure S17b, Supporting Information). These migrations by rolling were significantly faster than sliding because they were not affected by hydrodynamic resistance from the lubricant and friction by a large contact area; centimeter scale movements could be realized in a second. Smaller droplets moved faster due to the smaller mass (Figure 6f). Surprisingly, the smallest droplet of $2\text{ }\mu\text{L}$ droplet exhibited ultrafast motion of $\approx 10\text{ cm s}^{-1}$, which was orders of magnitude faster than in previous studies.^[46,81,82]

3. Conclusion

In summary, we present a MIR laser-powered droplet control strategy on liquid repellent surfaces. Localized droplet heating induced by the MIR laser (wavelength of $10.6\text{ }\mu\text{m}$) triggers thermocapillary convection to propel the droplet. Since liquid molecules on the outer surface of the droplet spontaneously generate heat due to their high absorption coefficient for MIR light, a powerful thermocapillary convection is produced within a fast response time. Therefore, local heating using the MIR laser enables highly efficient and precisely controllable droplet manipulation. Droplet motion by sliding is 2–5 times faster than manipulation methods based on photothermal additives. Notably, rolling motion on a superamphiphobic surface exhibits a velocity on the order of centimeters per second, which is orders of magnitude faster than sliding motion. Moreover, indirect droplet heating through a substrate, another advantage of the MIR laser, offers several advantages, including multi-droplet manipulation and droplet control under a cover glass. This method does not require photothermal additives, making it applicable to various liquid droplets and liquid repellent surfaces. Purification to remove additives is also unnecessary, which is advantageous for microreactor applications. Consequently, droplet manipulation using MIR laser irradiation will provide novel insights into droplet control with broad applications.

4. Experimental Section

Fabrication of the Lubricated Surface (LuS): Slide glass was washed sequentially with diluted Hellmanex III in water, ethanol, and acetone by ultrasonication for 15 min. Then, the slide glass was cleaned by using air plasma (PDC-002, Harrick, America) for 10 min. After cleaning, the slide glass with silicone oil (trimethylsiloxy-terminated PDMS, viscosity:

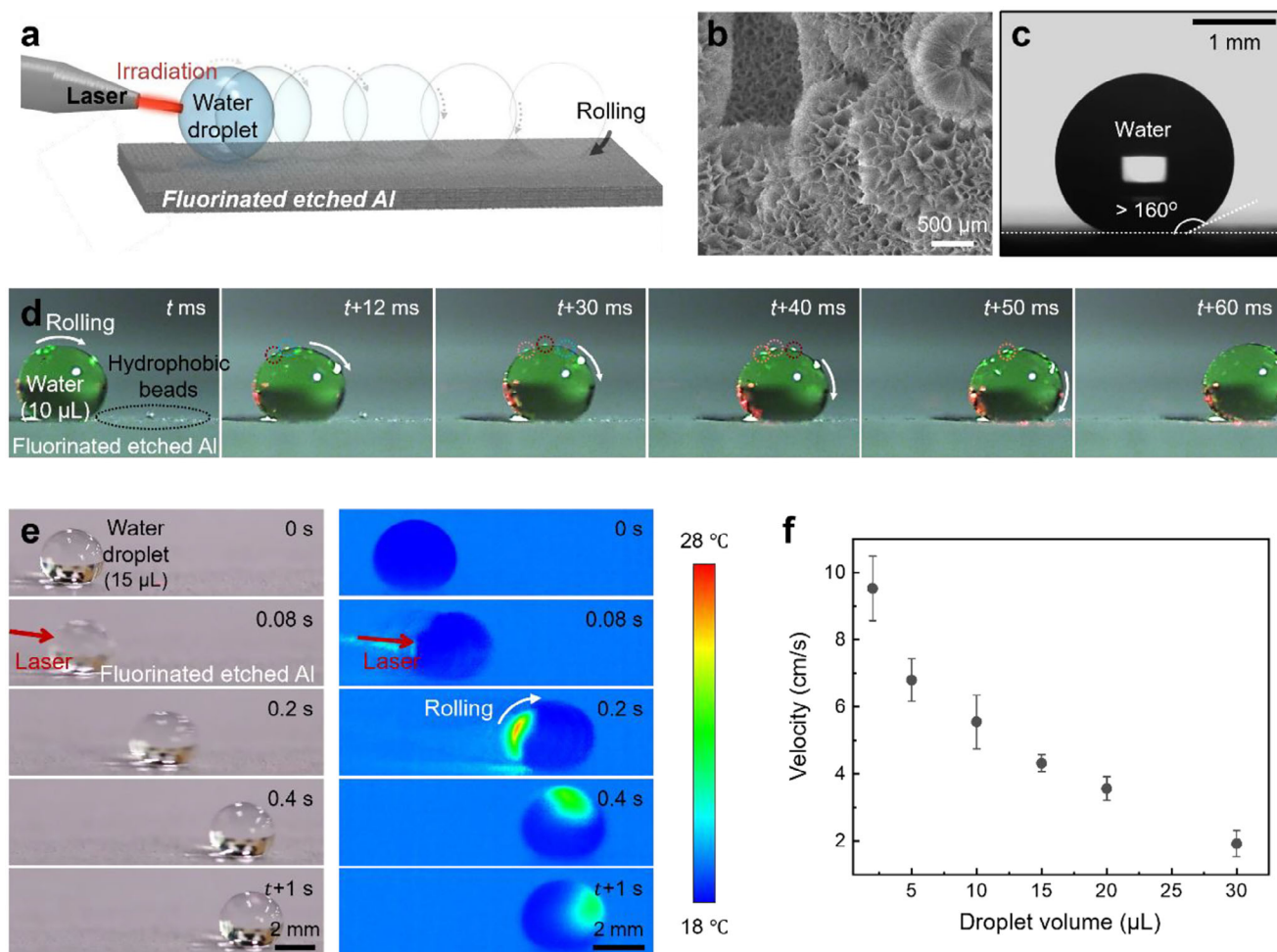


Figure 6. a) Schematic showing the rolling water droplet on the fluorinated etched aluminum by the laser. b) Surface image of the fluorinated etched aluminum by field emission scanning electron microscope (FE-SEM). c) The image represents the apparent water contact angle on the surface. d) Snapshots demonstrating the rolling water droplet (10 μL) passing over the hydrophobic beads captured by a high-speed camera. e) Pictures show the optical (left) and IR images (right) of droplet rolling by the laser intensity of 26.6 W mm^{-2} . f) Velocity of water droplets on the fluorinated etched aluminum depending on droplet volume.

100 cSt, Gelest Inc.) was stored in an oven at 100°C for 48 h to graft PDMS brush. Residual silicone oil after the grafting reaction was sequentially washed with hexane, toluene, and tetrahydrofuran. Finally, silicone oil (viscosity: 5 cSt, Gelest Inc.) was coated on the PDMS grafted surface by using a spin coater at 1000 rpm for 60 s.^[64]

Fabrication of the Superamphiphobic Surface: To demonstrate rolling motion, two different superamphiphobic surfaces were used in this study: i) etched aluminum- and ii) soot-templated surfaces, prepared following previous literature.^[76,77] To fabricate the etched aluminum-based superamphiphobic surface, an aluminum plate ($50 \text{ mm} \times 50 \text{ mm} \times 2 \text{ mm}$) was etched in a 2.5 M HCl solution at room temperature for 15 min to form microstructures. After etching, the plate was rinsed with deionized water and dried at 100°C for 15 min. The plate with microstructures was then placed in boiling water for 15 min to fabricate nanopores, followed by drying at 100°C for 10 min. The surface of the etched aluminum plate was rendered hydrophobic by chemical vapor deposition (CVD) with trichloro(1H,1H,2H,2H-perfluorooctyl)silane in a vacuum chamber for 2 h. Subsequently, it was stored in an oven at 100°C for 15 min to remove all unreacted reagents.

The soot-templated superamphiphobic surface was fabricated by depositing a soot layer onto the glass surface. The glass was placed above

the flame of a paraffin candle for 10 min, resulting in the formation of a black-colored soot layer on top of the glass. The soot-deposited glass was placed in a desiccator to form a silica shell ($20 \pm 5 \text{ nm}$) for 48 h via CVD with tetraethyl orthosilicate (400 μL) and ammonia (400 μL) under ambient conditions. Subsequently, the glass with the soot layer underwent calcination to combust the carbon core at 500°C for 4 h. Then, the substrate was fluorinated by utilizing the same CVD process as for the etched aluminum based superamphiphobic surface.

Fabrication of the Lubricant Impregnated Surface (LIS): To create the porous structure for the LIS, a mesoporous TiO_2 layer (thickness: $\approx 120 \text{ nm}$) was introduced.^[83] The TiO_2 layer was formed by sintering the doctor blade coated TiO_2 nanoparticle paste (18NR-T, Dyesol, diameter: $\approx 20 \text{ nm}$) at 500°C for 4 h. Afterward, this TiO_2 layer was incubated in silicone oil (viscosity: 100 cSt, Gelest Inc.) at 100°C for 48 h to graft PDMS brush on the surface. The substrate was sequentially washed with hexane, toluene, and tetrahydrofuran to eliminate residual oil. For lubricant impregnation, silicone oil (viscosity: 5 cSt, Gelest Inc.) was spin coated onto the PDMS brush grafted TiO_2 layer at 1000 rpm for 60 s.

Enzymatic Cascade Reaction Using Glucose Oxidase (GOX)/Horseradish Peroxidase (HRP): To conduct the glucose assay, a commercial enzyme mixture (4 mg GOX and 1 mg HRP) was dispersed in 1 mL of water, which

was used as an indicator solution. Analyte droplets, glucose aqueous solutions, were prepared with glucose with varying glucose concentrations (1, 20, 50, and 500 mM) and 50 mM KI.

Droplet Manipulation: A CO₂ laser (Finexel, SNJ, Korea) with a wavelength of 10.6 μm (power: 100–500 mW, beam size: 0.038 mm²) was used to propel liquid droplets on the liquid repellent surfaces. To investigate the droplet motion at certain velocities, a motorized stage (VT-80, PI, Korea) was employed. While monitoring droplet motions, the laser was fixed on the stage and moved at a given velocity. The stage speed was accelerated until the laser continuously irradiated the receding edge of the droplet.

Characterization of Surface Wetting Property: The wetting properties, including apparent contact angles, sliding angles, and contact diameters of various liquid droplets on the liquid repellent surfaces, were measured by using a contact angle measurement setup (DSA100, KRÜSS GmbH, Germany). Each measurement was repeated 5 times to obtain average values. The wetting ridge of the lubricant around the water droplet was observed by confocal microscopy (Leica Microsystems TCS SP8, Germany) with a dry objective 40 ×/0.85. The water droplet and silicone oil were dyed with Atto 488 and Lumogen Red, respectively. To capture the water droplet motion, a high-speed camera (Fastcam Mini AX100, Photron, Japan) which was capable of recording at up to 4000 frames per second (fps) at a resolution of 1024 × 1024 pixels. For our experiment, imaging were acquired at 1000 fps with an optical lens (AF-S Micro NIKKOR 105 mm f/2.8G ED, Nikon, Japan). Subsequently, the recorded videos were analyzed using ImageJ (National Institutes of Health).

Temperature Measurement: Temperature changes were observed by using the infrared camera (A655SC, FLIR, America) under laser irradiation. To measure temperature variation over time, the working distance between the laser and the droplets (or surfaces) was fixed at 3.5 cm. The emissivity of the IR camera was set to 0.95, a commonly adopted value for non-metallic materials such as water and glass.^[84,85] To prevent optical interference and damage to the MIR laser, a laser cut-off filter was placed in front of the IR camera, which blocks wavelengths above 10.2 μm and transmits over 90% of light below 10.2 μm (±5%).

Flow Analysis: To visualize internal flows at the top-view, the flow inside the droplet was observed by using an optical microscope. The flow from the side-view was monitored with a camera for the contact angle measurement setup. For these measurements, polystyrene and hydrophobized TiO₂ particles were used as tracer particles.^[86]

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Author Contributions

S.W. and H.H. designed the research. H.H. and J.L. conducted experiments. H.H. analyzed data. Y.Z., K.L., and C.L. performed simulation analysis. All authors have discussed the results. The manuscript was written through the contributions of all authors. All authors have given approval to the final version of the manuscript.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

droplet manipulation, liquid repellent surface, Marangoni flow, MIR irradiation, thermocapillary convection

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