Liquid Metal Gallium Micromachines Speed Up in Confining Channels

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One of the great challenges in the field of micro- and nanotechnology is the capability of engineering liquid metal micromachines to autonomously move within confining channels to perform various complex tasks. Herein, liquid metal gallium micromachines that significantly increase their velocity in confining channels with adaptive deformation under exposure to an electric field are presented. The liquid metal gallium micromachines move toward the negative electrode under the propulsion of hydrogen bubbles, which is obviously different from the previous report that liquid metal gallium alloy (i.e., Galinstan) micromachines move to the positive electrode, owing to the surface tension gradient. More importantly, the liquid metal gallium micromachine can adaptively deform and accelerate in confined channels. The velocity of liquid metal gallium micromachines increases with the decrease in the width of channels. It is found that the speed-up motion of liquid metal gallium micromachines is caused by the enhanced electro-osmosis effect in confining channels. The findings not only help understand the role of the confinement effect on the motion of liquid metal micromachines but also provide a novel strategy to manipulate the motion velocity of liquid metal micromachines for specific applications.

Micromachines capable of converting chemical energy and other forms of energies to mechanical movement have received much attention over the past decades. These micromachines hold promise in the fields of microfabrication, material assembly, detection, sensing, detoxification, and targeted drug delivery. Most of the current micromachines are composed of rigid materials, due to the limitation of deformation; however, it remains a challenge for the specific applications of the reported micromachines. As is well known, the liquid metal gallium and gallium-based alloys display fluidic and metallic properties and have deformability, a low melting point, and a supercooling effect at room temperature. With the release of the Hollywood film “Terminator 2,” more and more studies have focused on how to construct a liquid metal micromachine with a T-1000-like motion and deformation capability. Recently, several liquid metal-based micromachines were reported, which can autonomously move in fluids under the propulsion of bubble, pressure, ionic gradient, ultrasound field, electrical field, and magnetic field. To achieve the applications in complex environments, liquid metal micromachines are often required to move in confining channels. However, to our best knowledge, the influence of the confining environment on the motion of the liquid metal-based micromachine has not been reported yet, which is important for the precise control of their movement behavior and future applications.

Herein, we report that the liquid metal gallium droplet micromachines propelled by the generated hydrogen bubbles move toward the negative electrode in sodium hydroxide (NaOH) solution under exposure to an electric field, but they speed up with the adaptive deformation in the confining channels. The movement velocities of the gallium droplet micromachines increase with the width of the microfluidic channels. We demonstrate that this speed-up effect in the confining channels is ascribed to the increasing electro-osmotic effect. Our work provides a new proof for the underlying mechanism of electrically driven liquid metal micromachines and also their motion manipulation.

Figure 1A schematically illustrates the autonomous movement of the liquid metal Galinstan and gallium micromachines in the NaOH solution under a direct current (DC) electric field at 35°C. Liquid metal Galinstan and gallium micromachines with a mass of 50 mg were submerged, respectively, in a polydimethylsiloxane (PDMS) channel with dimensions of 12 cm long × 10 mm wide × 10 mm high, containing 1 M NaOH solution. After applying a DC electric field with a voltage of 30 V, the liquid metal Galinstan micromachine moved toward the positive electrode at a speed of 117.2 mm s⁻¹ (Figure 1B). The corresponding top and side images in Figure 1C demonstrate that the shape of the liquid metal Galinstan micromachine deformed during the movement. Interestingly, under the same conditions, the liquid metal gallium micromachine moved toward the negative electrode with a velocity of 2.8 mm s⁻¹ (Figure 1D). We observed that the liquid metal gallium micromachine not
only changed its shape but also generated a great number of bubbles on the opposite movement (Figure 1E).

It is previously reported that the motion of the liquid metal Galinstan micromachine is propelled by the surface tension gradient.\textsuperscript{34–37} As long as external potential is applied, surface tension on the negative electrode side of both liquid metal Galinstan and gallium micromachines is larger than that on the positive electrode side. The only difference is that the liquid metal gallium micromachine can continuously generate hydrogen bubbles, whereas the liquid metal Galinstan micromachine cannot. This is because the standard electrode potentials of indium (0.99 V) and tin (0.909 V) are lower than that of gallium (1.219 V), the oxidation of indium and tin is preferred in the NaOH solution. As the oxide layer of indium and tin cannot be dissolved by NaOH, which would block the reaction between gallium and NaOH, the formation of hydrogen bubbles was impeded. In contrast, because gallium oxide can be dissolved in NaOH solution, hydrogen bubbles can be continuously generated by gallium micromachines. Also, the electrode reactions of the liquid metal gallium micromachine are expressed as

\[ \frac{4}{3} \text{Ga} + \frac{16}{3} \text{OH}^- \rightarrow \frac{4}{3} \text{[Ga(OH)]}_4^- + 4e^- \] (1)

\[ 2\text{H}_2\text{O} + 4e^- \rightarrow 2\text{H}_2 \uparrow + 4\text{OH}^- \] (2)

Therefore, we speculate that the motion of gallium micromachines is propelled by hydrogen bubbles. To verify the motion mechanism of the liquid metal gallium micromachine, isopropanol (IPA), as a defoaming agent,\textsuperscript{38} was added into the NaOH solution and the motion velocities of liquid metal gallium and Galinstan micromachines were calculated. As shown in Figure 1F, the velocity of gallium micromachines decreased with the increase in the concentration of IPA, while there was no significant change in the velocity of Galinstan micromachines before and after adding IPA. These results demonstrate that the motion of gallium micromachines toward the negative electrode is mainly driven by the generated hydrogen bubbles.

To investigate the motion behavior of liquid metal gallium micromachines in the confining channels, a gallium droplet micromachine with a mass of 50 mg and an initial diameter of 2.5 mm was dropped into an open-up PDMS channel with five sections including section “a” and “e” (2 mm wide × 30 mm long × 10 mm high), section “c” (1 mm wide × 30 mm long × 10 mm high), and intersection “b” and “d,” as schematized in Figure 2A. The time-lapse images in Figure 2B (captured from Video S1, Supporting Information) illustrate the movement of a liquid metal gallium micromachine within the aforementioned PDMS channel containing 1 mol L\textsuperscript{-1} NaOH solution under a 30 V DC field. It was found that the gallium micromachine moved from the positive electrode to the negative electrode, which is consistent with the result in Figure 1D. The corresponding top-view images in Figure 2C demonstrate that the motion of the gallium micromachine is accompanied by the continuous generation of hydrogen bubbles. More importantly, the liquid
metal gallium micromachine shows adaptive deformation in the process of movement. The deformation degree of gallium micromachines increased with successive decrease in the width of the channel.

To further investigate the movement of liquid metal gallium micromachines under confinement, we measured their motion velocity and the degree of deformation. The deformation degree was calculated as \( \frac{L}{L_0} \), where \( L \) is the real-time length of the liquid metal micromachine along the direction of the channel and \( L_0 \) is the initial diameter of the liquid metal micromachine outside of the channel. As shown in Figure 2D, the deformation degree of liquid metal gallium micromachine initially increased and then decreased with the changing of the width of channels. It can be found that the deformation degree of liquid metal gallium micromachine in the channel section “a” was 1.42, similar to that in the section “e” (1.45). In the channel section “c,” the deformation degree was 1.65 times of that in the channel section “a.” These results demonstrate that the liquid metal gallium micromachine can adapt to the channel width, and the deformation degree of the gallium micromachine depends on the width of the channel. The instantaneous velocity of the liquid metal gallium micromachine exhibited a similar trend with the deformation degree in the confining channel (Figure 2E). In particular, the motion velocity in channel section “a” was 8.6 mm s\(^{-1}\) which is about 3.1 times of that in Figure 1D. We also found that the motion velocity increased rapidly as the width of the intersection channel “b” decreased. After the gallium micromachine completely entered channel section “c,” motion velocity increased to 23.9 mm s\(^{-1}\), which is 2.8 times of that in channel “a.” When the gallium micromachine swims from channel section “c” to “d” and “e,” its motion velocity decreases gradually to its initial values. Taken together, these results demonstrate that the liquid metal gallium micromachines can adaptively deform and speed up in confining channels, and the degree of deformation and the velocity of motion are related to the width of the confined channels.

Based on the dynamic performance of liquid metal gallium micromachines in the channels, under certain condition of the electric field, the factors that determine the speed-up motion of the gallium micromachines are the concentration of the

**Figure 2.** Liquid metal gallium micromachines speed up in confining channels under a 30 V DC electric field. A) A schematic representation of the movement of the gallium micromachine in a microfluidic channel with different section widths. The letters a, b, c, d, and e represent different channel sections. B) Time-lapse images of the gallium micromachine with a mass of 50 mg in an open-up PDMS channel. The width of the channel section: “a” and “e” is 2 mm; “c” is 1 mm. C) Top-view images of the motion of the gallium micromachine in each channel section. D) The relationship of the deformation degree of the gallium micromachine and its position in the channels. E) The instantaneous velocity analysis of the gallium micromachine in the process of movement in (B). Scale bars: 5 mm (B); 1 mm (C).
NaOH solution, the initial diameter of the gallium micromachines, the width of the channels, and, where the initial diameter depends on the mass of the gallium micromachine. First, by studying the motion behavior of the liquid metal gallium micromachine in channels containing NaOH with different concentrations, we found that the movement velocity of the liquid metal gallium micromachine increased with the increase in the concentration of NaOH solution and the applied electric voltage (Figure S1, Supporting Information). To study how confinement influences liquid metal gallium micromachine dynamics and reveals the mechanism of the speed-up motion, we investigated the dependence of motion velocity on the mass of the gallium micromachine. As shown in Figure 3A, under the same electric field, the velocity of the gallium micromachine increased with the increase in the mass of the micromachine in the 1 mm-wide channel with 1 m NaOH solution. A similar trend was observed in the channel with a width of 2 mm, where the velocity also increased with the mass of the gallium micromachine (Figure 3B). Then, the effect of the channel width on the velocity of the gallium micromachine was studied. Gallium micromachines with a mass of 50 and 80 mg were submerged, respectively, in channels with a width of 1, 2, and 3 mm. It was observed that the velocity of the gallium micromachine decreased with the increase in the channel width under a certain electric field (Figure 3C,D).

Theoretically, the adaptive deformation and speed-up motion of the liquid metal gallium micromachine in the confining channel are governed by the bubble thrust force $F_b$, surface tension force $F_\gamma$, electro-osmotic force $F_e$, friction force $F_r$, and viscous force $F_\eta$ from the solution (Figure 3E). These forces are related by the energy balance equation as

$$\left(F_b + F_e - F_r - F_\gamma - F_\eta\right) \cdot \Delta x = \frac{1}{2} \rho_m V (v^2 - v_0^2) + \frac{1}{2} K (L^2 - L_0^2)$$  \hspace{1cm} (3)

where the $\Delta x$ and $K$ are the displacement and elastic coefficient of the liquid metal micromachine and $L$ is the length of the micromachine. As the gallium micromachine enters the narrow channel, the velocity and length of the micromachine increase, indicating that the net force $(F_b + F_e - F_r - F_\gamma - F_\eta)$ also increases with the same displacement.

To understand the mechanism of the speed-up motion, we first evaluated the change of $F_\gamma$. It is assumed that the electric charge is distributed uniformly on both hemispheres of the gallium micromachine; thus, the pressure difference ($P$) of the liquid metal micromachine can be described by Young–Laplace’s equation

$$P = \frac{\gamma}{r}$$  \hspace{1cm} (4)

where $\gamma$ is the surface tension and $r$ is the curvature radius. To simplify the calculation, the curvature radii of the two ends of the micromachine are considered equal, which corresponds to half of the width of the channel ($w$). Also, the pressure difference can be expressed as

![Figure 3. The mechanism analysis of the speed-up motion of the liquid metal gallium micromachine in the confining channels. The velocity curve of the gallium micromachine with different masses in straight channels with widths of A) 1 mm and B) 2 mm under an electric field with different voltages. The average velocity of the gallium micromachine with a mass of C) 50 mg and D) 80 mg in straight channels with different widths under electric fields. E) A schematic representation of the forces affecting the velocity of the gallium micromachine. F) The illustration of electro-osmosis effect on the gallium micromachine.](image-url)
\[ \Delta P = P_R - P_L = (\gamma_R - \gamma_L) \frac{4}{w} = \frac{4\Delta \gamma}{w} = \frac{4q_b \Delta \varphi}{w} \quad (5) \]

where \( q_b \) is the initial surface charge density and \( \Delta \varphi \) is the potential drop across the liquid metal micromachine. \( \Delta \varphi \) can be evaluated as

\[ \Delta \varphi = \frac{V_f R_M}{R_T} \quad (6) \]

where \( V_f \) is the applied voltage, \( R_M \) is the resistance of the electrolyte solution between the liquid metal micromachine and the PDMS wall, and \( R_T \) is the total resistance. The resistance can be expressed separately as

\[ R_M = \rho_e \frac{L}{S} - \frac{\rho_e L^2}{hwL - V} \quad (7) \]

\[ R_T = R_M + R_e = \frac{\rho_e L^2}{hwL - V} + \frac{\rho_e (d - L)}{hw} \quad (8) \]

where \( \rho_e \) is resistivity of the electrolyte solution, \( S \) is the area of the cross section, \( h \) is the height of the electrolyte solution (10 mm), \( V \) is the volume of the micromachine, and \( d \) is the distance between the two electrodes (10 cm). The force induced by the surface tension gradient of the droplet can be derived from Equation (4)–(7) as

\[ F_{\gamma} = \Delta \varphi \pi r^2 = \pi q_b V_f \frac{hL^2w^2}{hwLd + LV - dV} \quad (9) \]

For the liquid metal gallium micromachine with a mass of 50 mg, the value of \( L \) was 3.6 and 5.9 mm in the channel with a width of 2 and 1 mm, respectively. It can be found that \( F_{\gamma} \) increased when the liquid metal gallium micromachine entered the narrower channel.

The viscous force \( F_{v} \) could also be estimated based on the following formula

\[ F_v = 6\pi \eta \rho v k_1 k_2 \quad (10) \]

where \( \eta \) is the viscosity of the electrolyte, \( v \) is the velocity of the micromachine, and \( k_1 \) and \( k_2 \) are the correction factors for the shape of the liquid metal micromachines. It can be found that \( F_v \) increased with the increase in the velocity of the gallium micromachine, indicating the increased resistance in the narrow channel. Meanwhile, the friction force \( F_f \) also increased with the increase in pressure after the liquid metal gallium micromachine enters the narrow channel. For this system, the bubble thrust force \( F_b \) is regulated by the voltage of the external electric field and electrolyte concentration. As the gallium micromachine moved from a wide channel into a narrow channel, \( F_b \) was stable at constant voltage and ion concentration.

Taken together, it can be inferred that \( F_v \) increased as the gallium micromachine enters the narrow channel and dominates the acceleration of the micromachine. This is mainly because the thickness of the slip layer \( h^{(10)} \) between the gallium micromachine and PDMS channel wall decreases with the increase in the confining effect, leading to the enhancement of electro-osmosis, as schematically shown in Figure 3F. Taken together, the speed-up characteristic of the liquid metal gallium micromachine comes from the increased electro-osmotic force.

In conclusion, we have demonstrated the adaptive deformation and speed-up motion of the liquid metal gallium micromachine in the confining channels upon applying DC electric fields. Compared with the liquid metal Galinstan micromachine, which is propelled by the surface tension gradient to move toward the positive electrode, the liquid metal gallium micromachine moves to the negative electrode under the propulsion of the generated hydrogen bubbles. More importantly, the liquid metal gallium micromachines exhibit a shape-transformable movement in the confining channels, and their motion velocities increase with the decrease in the width of the channels. The experimental results reveal that this speed-up motion of the liquid metal gallium micromachine in the confining channel is attributed to the enlarged electro-osmotic effect. Such a liquid metal gallium micromachine represents a novel micromachine with a similar deformation and movement capabilities as T-1000 and provides a new possibility for the motion control of the micromachine in the channels.

**Experimental Section**

**Materials:** Gallium, Galinstan, NaOH, and IPA were purchased from Aladdin. Sylgard 184 PDMS and the curing agent were obtained from Shanghai Haitong Trading Co., Ltd. Graphite electrodes (Φ3 × 20 mm) were purchased from Beijing electric carbon factory. The alligator clip wires were purchased from the Imagination teaching instrument. The APS3003S DC power supply was purchased from Shenzhen ATTEM Technology Co., Ltd.

**Fabrication of PDMS Channel Device:** The open-top PDMS channels were fabricated using standard soft lithography. To fabricate the PDMS channels, a PDMS substrate was prepared first by mixing a silicone- elastomer base and curing agent at the ratio of 10:1 by weight. Then, the mixture was poured into a glass container, followed by degassing in a vacuum oven. After that, the molds were placed on the substrate and completely covered using the PDMS substrate. After curing at 80°C for 2 h, the molds were separated carefully from PDMS with tweezers. Also, the PDMS channels were obtained by peeling off from the glass container.

**Actuation of the Liquid Metal Micromachines:** To actuate the liquid gallium and Galinstan micromachines, the as-prepared PDMS channel was filled with 1 M NaOH solution. After that, two graphite electrodes were placed in the reservoirs at a distance of 10 cm. To maintain the experiment temperature at 35°C, the PDMS channels were placed on a heating plate. Then, the propulsion experiments were performed by separately injecting liquid metal gallium and Galinstan micromachines into the reservoir of the positive electrode and applying a DC electric field. The movement of the liquid metal micromachines was observed and recorded using a digital camera.

**Optical Image Observation:** To record the morphology of gallium and Galinstan micromachines before and after the application of the DC electric field, an optical microscope (Olympus DP72) was used to capture the top-view images of micromachines. Also, the side-view pictures of the liquid metal micromachines were imaged by the side microscope of contact angle instrument (POWERAICH J2000D2).

**Velocity Analysis:** To calculate the velocity of the liquid metal micromachine, the movement of the liquid metal micromachine was observed and recorded using a digital camera. Then, the movement of liquid metal micromachine was analyzed using the open-source software ImageJ. The velocity of the micromachine was calculated by measuring the trajectory length. For each experimental condition, at least five micromachines were averaged.

**Supporting Information**

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

This work is financially supported by the National Natural Science Foundation of China (No. 21573053) and National Postdoctoral Program for Innovative Talents (BX201700065).

Conflict of Interest

The authors declare no conflict of interest.

Keywords

electro-osmosis, gallium, liquid metals, micromachines, speed up

Received: June 30, 2019
Revised: July 20, 2019
Published online: September 6, 2019

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