Motion Enhancement of Spherical Surface Walkers with Microstructures

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Surface walkers are widely studied as carriers for biomedical applications, such as targeted drug delivery and cell manipulation. For in vivo/in vitro therapeutic applications, high movement velocity is necessary for efficient operation on targets with high throughput. Herein, a fast underwater microstructured spherical surface walker (i.e., microsphere) driven by a rotating magnetic field is reported, and the effect of the surface microstructures on the mobility of microspheres is explored. Compared with the motion of smooth sphere walking nearby a smooth plane, a twofold and a fourfold increases in the velocity are found for the microstructured sphere (MS) walking nearby a smooth plane and a patterned plane, respectively. A hydrodynamic model of MS moving nearby a plane is used to reveal the underlying mechanism of the enhanced motion, which demonstrates that the slippage of fluid on the microstructured surface and the interaction between fluid and microstructures are crucial to the motion enhancement. The result of motion enhancement induced by microstructures can be used for the design of fast biomimetic microrobots, anti-adhesion, cell manipulation, and targeted drug delivery in complex aqueous environments.

1. Introduction

At the microscale in aqueous environment, symmetrical micro-objects will perform a reciprocal motion without any net displacement, because viscous forces predominate over inertial forces at low Reynolds number ($Re < 1$), which is referred to as “scalloping theorem.”[31] Inspired by micro-organisms, such as bacteria and cell,[2,3] introducing a nearby wall or asymmetric structures to the micro-objects can break the flow symmetry and, thus, realize an effective motion,[4-7] which is of benefit to the design of underwater microrobots.[8-19] “Surface walker” is the typical microrobot that utilizes the nearby wall to achieve the non-reciprocal motion, such as microspheres,[20-27] microwheels,[28-30] colloidal rotors,[31-33] and microrobot swarms.[34-37] They are potentially used in various biomedical applications, including targeted drug delivery and minimally invasive surgery,[38] in which a confined wall such as blood vessels and microchannels is naturally presented to facilitate the net motion of surface walkers.[19]

Motion performance of surface walker is crucial to the biomedical applications as a high translational velocity enables efficient operations on the target with high throughput.[39–42] There are various factors that influence the translational velocity of surface walker, such as the slippery properties of the confining wall (slip or no-slip boundary conditions),[24,42,43] the nature of the surface walker (e.g., the geometry and size of the surface walker),[44,45] the distance between the surface walker and the nearby wall,[21] and the actuation (e.g., magnetic field and frequency).[30–32] Taking spherical surface walker (i.e., microsphere) as an example, microspheres have a higher translational velocity on plane surfaces with micro-wells and micropillars or on slip wall,[42,43] and when multiple microspheres are combined into a whole with different shapes and sizes, their velocity will be greater than a single microsphere.[30–32] Besides, the translational velocity of microspheres increases as the applied magnetic frequency increases when lower than the step-out frequency.[30,31,33]

Apart from these factors, the surface microstructures as an important geometric factor have attracted much attention as they are usually distributed on the surface of the micro-organisms and play an important role in the propulsion and movement of the micro-organisms.[1,4-6] Bacteria and cells have mastered highly effective means of microscale motion through the aid of the surface microstructures, exemplified by bacterial flagella and cellular cilia.[1,7,46] Cilia are found in virtually every cell. They are several to dozens of micrometers in length and hundreds

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The ORCID identification number(s) for the author(s) of this article can be found under https://doi.org/10.1002/aisy.202000226.

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DOI: 10.1002/aisy.202000226
of nanometers in diameter and can provide propulsion power for cell movement.\textsuperscript{[47–49]} To date, the propulsion of the microorganisms through the active deformation of surface microstructures has been investigated,\textsuperscript{[1–8,50–52]} such as the helical corkscrew propulsion mechanism for bacterial flagella and the traveling-wave propulsion mechanism for cellular cilia. However, the effect of the presence of surface microstructures themselves on motion has not been researched yet. Researches on the microstructured surface walkers are helpful to reveal the motion mechanism of living micro-organisms in aqueous environment.\textsuperscript{[7,8]}

In this work, the movement performance of a spherical surface walker (i.e., microsphere) with surface microstructures (i.e., micropillars) has been investigated. The spherical shape is chosen, because it is very universal in micro-organisms. The microspheres are fabricated by 3D direct laser writing (DLW), which enables the manufacturing of custom-shaped microparts by various materials.\textsuperscript{[53–57]} The microspheres are propelled with a rotating magnetic field, because it can offer a remote, fuel-free, and engineless propulsion in a variety of fluidic environments.\textsuperscript{[8–12,58]} The smooth sphere (SS) and microstructured sphere (MS) are propelled to translate not only near a smooth plane but also a patterned plane with micropillars, because the morphology of the plane surface plays an important role in determining the nature of boundaries.\textsuperscript{[59–65]} The experimental results show that compared with an SS translating near a smooth plane, an MS can perform a 2.2 and 4.1 times increase in translation velocity when translating near a smooth plane and a patterned plane, respectively. Through theoretical analyses, this motion enhancement is ascribed to the slippage of the microstructure surface and the interaction between fluid and microstructures due to fluid flowing in and out of the microstructures.

2. Results and Discussion

2.1. Design and Fabrication

The design of the SS and the MS is shown in Figure 1a,b, respectively. The MS consists of an inside solid sphere with
radius $R_m$ and an outer micropillar array with pillar height $h$, pillar diameter $d$, and pillar quantity $n$. The radius of the entire MS is equal to the radius of the SS, $R$, i.e., $R = R_m + h$. In this work, $R = 40 \, \mu m$, $h = 10 \, \mu m$, $d = 4 \, \mu m$, and $n = 128$, and the solid fraction of the micropillars on the sphere surface is $\phi = nd^2/(16R^2) = 0.08$. The morphology of the microsphere is well designed to match the scale of paramecium, a common single-celled protozoan. The paramecium usually has a body length from 80 to 300 $\mu m$ and the size of the cilium on the surface of paramecium is around 8 $\mu m$ in length and 0.5 $\mu m$ in diameter. The fabrication process of microspheres is shown in Figure 1c. Magnetic microspheres were fabricated by 3D DLW with SU-8 photoresist, followed by the selective physical vapor deposition of a 150 nm thick nickel film on the sphere surface. Figure 1d,e shows the SEM images of the SS and the MS, respectively. The microspheres were removed from the sample substrate and transferred to the microtank to implement the motion experiments using a sharp probe controlled by a micro-manipulator. In addition, the patterned plane was fabricated by the lithography and etching process on a silicon substrate to have a micropillar array with the diameter of 2 $\mu m$ and the cover fraction of 19.6%, as shown in Figure 1f,g.

### 2.2. Movement Performance

To explore how surface microstructures affect the motion behavior of magnetic spherical surface walkers, an MS was used to move on the smooth plane and patterned plane in deionized (DI) water in the presence of an in-plane rotating magnetic field. Initially, the water is still. Identical experiments were performed with an SS as the control experiment.

#### 2.2.1. Movement of Microspheres on Smooth Plane

First, we experimentally studied the movement performance of MS and SS moving on smooth plane to investigate the effect of microstructures on the velocity of the microspheres. The motion images of the MS and the SS on the smooth plane are shown in Figure 2a, in which the magnetism-driven frequency is 10 Hz, and the magnetic field strength is 14 mT. The forward translating velocity of the MS is $267 \, \mu m/s$, whereas the SS is $94 \, \mu m/s$, indicating that the MS exhibits a better movement efficiency than the SS (see Movie S1, Supporting Information, in which the magnetism-driven frequency is 1 Hz, and the magnetic field strength is 14 mT). The variations of the forward velocities of microspheres as a function of the input rotational frequency of the magnetic field at 14 mT are shown in Figure 2c. Each data point in Figure 2c represents an average of six trials characterized from three different microspheres. As shown in Figure 2c, the forward velocities of microspheres exhibit an approximately linear relationship against the input rotating frequency when the sphere rotates in sync with the input magnetic field, and then reach the peak at the step-out frequency (i.e., the frequency corresponding to the maximum velocity). When the input rotational frequency of the applied magnetic field is higher than the step-out frequency, increasing the frequency will gradually reduce the velocity, because the available magnetic torque is no longer sufficient to keep the rotation of the sphere synchronized with the applied field. For the SS, the measured step-out frequency is 6 Hz, and the corresponding maximum velocity is $\approx 123 \, \mu m/s$. For the MS, the measured step-out frequency is 10 Hz, and the corresponding maximum velocity is $\approx 267 \, \mu m/s$. The forward translation velocity of the MS is higher than that of the SS across the entire frequency range, and the maximum velocity of the MS is 2.2 times higher than the SS. This indicates that the introduction of surface microstructure can increase the forward velocity of the spherical surface walkers.

#### 2.2.2. Movement of Microspheres on Patterned Plane

Second, we experimentally investigated the movement performance of MS and SS translating on patterned plane. The motion
images of the MS and the SS moving on the patterned plane are shown in Figure 2b, in which the magnetism-driven frequency was 11 Hz, and the magnetic field strength was 14 mT. The forward translating velocity of the MS is 507 μm s⁻¹, whereas the SS is 227 μm s⁻¹. Similar to the cases of smooth plane, the MS performs a better movement efficiency than the SS (see Movie S2, Supporting Information, in which the magnetism-driven frequency is 9 Hz, and the magnetic field strength is 14 mT).

As shown in Figure 2c, the measured maximum velocities for the MS and the SS moving on the patterned plane are 507 μm s⁻¹ at 11 Hz and 271 μm s⁻¹ at 9 Hz, respectively. Furthermore, the velocity of the MS on the patterned plane is much higher than that of the SS on the smooth plane in the entire frequency range and even increases by 4.1 times for the maximum velocities. The result indicates that the movement efficiency of the spherical surface walker can be further enhanced by the introduction of the structures to the nearby plane.

2.3. Theoretical Model for Movement of Microspheres

2.3.1. Movement of Magnetism-Driven Microspheres in a Low Reynolds Number Flow

To reveal the underlying mechanism of motion enhancement when microstructures are introduced, a theoretical model is proposed to investigate the movement of an MS near a plane. As shown in Figure 3a,b, in an aqueous environment without incoming flows, the microsphere rotates on the plane with an angular velocity Ω and translates with a forward velocity U parallel to the plane under the action of external magnetic moment T_m.

For the low Reynolds number (Re) flow, the N–S equation and the continuity equation are expressed as

\[ \nabla p = \mu \nabla^2 u; \nabla \cdot u = 0 \]  
\[ \begin{align*}
F &= 6\pi \mu R F^\prime U + 6\pi \mu R^2 F^\prime r \Omega \\
T &= 8\pi \mu R^2 T^\prime U + 8\pi \mu R^3 T^\prime r \Omega \end{align*} \]

where \( F \) and \( T \) are the resistive force and torque from the fluid, respectively, \( F^\prime \) and \( T^\prime \) are the normalized scalar force and torque components. For an SS moving near a smooth plane, they can be expressed as \( F^\prime = (8/15) \ln(\delta/R) + O(\delta/R)^0 \), \( F^\prime r = -(2/15) \ln(\delta/R) + O(\delta/R)^0 \) and \( T^\prime = -(1/10) \ln(\delta/R) + O(\delta/R)^0 \), \( T^\prime r = (2/5) \ln(\delta/R) + O(\delta/R)^0 \) [21] where the superscripts “t” and “r” represent the components of the translation and the rotation of the sphere, respectively, \( \delta \) is the gap between the plane and the bottom of the sphere.

As shown in Figure 3a, b, in the presence of an external rotating magnetic field, the external magnetic moment \( T_m \) is exerted...
on the microsphere to align its magnetization, which is given by

\[ T_m = V|\mathbf{M} \times \mathbf{B}| = V|\mathbf{M}| |\mathbf{B}| \sin \theta \]  

(3)

where \( V \) is the volume of the Ni film sputtered on the microsphere, \( \mathbf{M} \) is the magnetization of the microsphere, \( \mathbf{B} \) is the magnetic field vector, and \( \theta \) is the angle between \( \mathbf{M} \) and \( \mathbf{B} \).

When subjected to the external magnetic moment \( T_m \), the uniform rolling of the microsphere forward along the plane can be regarded as a series of equilibrium processes of external magnetic moment \( T_m \) and the fluid resistance torque \( T \), that is, \( T_m = T \). In our case, the microsphere is only subjected to the external magnetic moment \( T_m \) without any magnetic force, indicating that \( F = 6\pi \mu R F^r U + 6\pi \mu R^2 F^r \Omega = 0 \) due to the force balance. Therefore, the relationship between \( U \) and \( \Omega \) can be expressed as

\[ \frac{U}{\Omega} = \frac{RF^r}{F^r} \]  

(4)

In addition, the maximum value of the magnetic moment is \( T_m = V|\mathbf{M}| |\mathbf{B}| \), and the corresponding rotational frequency \( \Omega \) is called the step-out frequency \( \Omega_{\text{step-out}} \). Thus, according to \( \Omega_{\text{step-out}} = 2\pi f_{\text{step-out}} \) and Equation (2) and (4)

\[ \Omega_{\text{step-out}} = \frac{F^r}{8\pi \mu R^3 (F^r T^r - F^r T^r)} T_{\text{max}} \]  

(5)

Normally, at a certain magnetic rotating field, the magnetic moment offsets the fluid resistance torque, and the microsphere swims steadily with a constant forward velocity where the rotational frequency of the microsphere synchronizes with the magnetic rotational frequency \( f \); in other words, \( \Omega = 2\pi f \). In this case, \( U \) shows an approximately linear relation against \( \Omega \) when \( \Omega < \Omega_{\text{step-out}} \). For \( \Omega = \Omega_{\text{step-out}} \), \( U \) and \( \Omega \) reach the maximum. Further increase in \( f \) results in the increase in the resistance torque on the microsphere while the magnetic moment stops increasing, which leads to a torque imbalance on the microsphere. Therefore, both the forward and the angular velocities of the microsphere will decrease to rebalance the resistance torque and the external magnetic moment, which is consistent with the experimental results in Figure 2c.

2.3.2. Hydrodynamic Model of MS

In this section, a theoretical method is developed to investigate the motion of microsphere near a patterned plane under the consideration of surface microstructures. According to Equation (4), to reveal the underlying mechanism of the motion enhancement, the normalized scalar force components \( F^r, F^r \) should be obtained by hydrodynamic analysis of the microspheres. Therefore, an analytical model derived from the fundamental equations of fluid mechanics is established under the consideration of the surface microstructures. As Equation (1) is linear, the motion of an MS on a patterned plane can be regarded as a superposition of translation and rotation.

Translation of MS: First, we consider an MS translating without rotation (\( \Omega = 0 \)) nearby a patterned plane. Following Goldman et al.,\(^{[21]} \) a cylindrical coordinate system (\( \rho, \phi, z \)) is used, as shown in Figure 3c. Coordinate origin is on the plane wall (denoted as P), and the z-axis passes through the sphere center. The sphere surface is denoted as S. The minimum gap is \( \delta = h - R \). Thus, Equation (1) can be expressed as \( \nabla p = \mu \nabla^2 U, \nabla \cdot U' = 0 \), where superscript “\( \prime \)” represents the translation (without rotation) of the sphere.

On the surface of the SS without slip, the velocity boundary condition is \( (u_\rho', u_\phi', u_z') |_S = (U \cos \phi, - U \sin \phi, 0) \), at which the fluid velocity will be equal to the motion velocity of microsphere.\(^{[21]} \) Here, the superscript “\( s \)” represents the SS. However, the presence of the microstructures on the sphere will affect the surrounding flow field. First, by introducing the microstructures, as the liquid wets the microstructures, the boundary of the microsphere is not a pure liquid-solid interface but can be regarded as a complex interface integrated with liquid-liquid and liquid-solid interfaces. Thus, the flow in the tangential direction will experience a slip boundary condition with a slip velocity \( U_s \), which influences the tangential velocity component \( u_\phi' \). Therefore, at the boundary, the fluid velocity will be smaller than the motion velocity of microsphere. Second, the fluid flows in and out of the microstructures and, thus, interacts with the microstructures, leading to the effect on the radial velocity component \( u_\rho' \). Hence, the velocity boundary condition on an MS is different from that on an SS. Here, we introduce parameters \( \alpha \) and \( \beta \) to denote the effect of the slippage of the microstructure surface on the tangential velocity component and the effect of the interaction between fluid and microstructures on the radial velocity component, respectively. In addition, the fluid will slip on the patterned plane with a slip velocity of \( U_s \). Therefore, for an MS translating on a patterned plane, as shown in Figure 3c, the velocity boundary conditions on sphere surface (S) and patterned plane (P) can be expressed as

\[
\begin{align*}
\left( u_\rho', u_\phi', u_z' \right) |_S &= (\beta U \cos \phi, - \alpha U \sin \phi, 0) \text{ on } S \text{ (sphere surface)} \\
\left( u_\rho', u_\phi', u_z' \right) |_P &= (U_p \cos \phi, - U_p \sin \phi, 0) \text{ on } P \text{ (plane)}
\end{align*}
\]

(6)

where the superscript “\( m \)” represents the MS. \( \alpha = 1 \) represents the case without slippage, and \( \beta = 1 \) represents the case without radial flow in and out of the surface microstructures.

To facilitate the investigation of the velocity distribution of the flow field on the microsphere surface in the \( x-y \) plane, as shown in Figure 3d, we convert the velocity components from the cylindrical coordinate system to the rectangular coordinate system, leading to \( u_\rho = (\beta - \alpha) U \cos \phi \sin \theta \). Meanwhile, the fluid flowing in and out of the microsphere is examined in the spherical coordinate system, in which the velocity of the fluid relative to the sphere in the radial direction is

\[ u_r = (\beta - 1) U \cos \phi \sin \theta \]  

(7)

Therefore, the fluid flowing in and out of the surface structures satisfies the mass conservation, i.e., \( \int_0^{2\pi} \int_0^\pi u_r \rho d\rho d\phi = 0 \). According to Equation (7), the smaller the \( \beta \), the larger the velocity of flow in and out of the microsphere, which reflects
a stronger interaction between the fluid and the moving microsphere.

Physically, when the sphere translates forward with a velocity of $U$, the fluid flows forward around the surface of the sphere. According to Van Dyke, when a sphere is falling steadily down along the axis of a tube with diameter twice as that of the sphere at $Re = 0.1$, the streamlines on sphere surface are left–right symmetry as well as up–down symmetry.[16,17] Therefore, at low $Re$, the projection of streamlines around the MS surface on the $x$–$y$ plane is assumed to be symmetric, as shown in Figure 3d. In Figure 3d, in the first quadrant of the sphere surface ($0 < \phi < \pi/2$), there should be $u_{\phi} = (\beta - \alpha) U \cos \phi \sin \phi < 0$, indicating that $\beta < \alpha$. Similarly, it is found that $\beta < \alpha$ is also valid in the second, third, and fourth quadrants.

In addition, as the tangential velocity components on the MS surface and the MS surface are $u_{\phi}|_S = \alpha U \sin \phi$ and $u_{\phi|^3}_S = \alpha U \sin \phi$, respectively, the tangential velocity difference between the presence and the absence of surface microstructures is ascribed to the slip boundary condition. Thus, we can obtain the relationship

$$U \sin \phi = \alpha U \sin \phi + U_s$$

indicating that $\alpha \leq 1$. According to Equation (8), the smaller the $\alpha$, the larger the $U_s$, resulting in a stronger slippage. Therefore, parameters $\alpha, \beta$ need to meet

$$\alpha \leq 1; \beta < \alpha$$

Following Goldman et al.[21] by solving Equation (1) with boundary condition (6) using a lubrication theory and singular perturbation method, the dimensionless force can be obtained (see details in the Supporting Information)

$$F^r = \left[\frac{4(\alpha + \beta)}{15} - \frac{8}{15} \frac{U_p}{U} \right] \ln \left(\frac{\delta}{R}\right) + O\left(\frac{\delta}{R}\right)^0$$

where $O(\delta/R)^0$ represents the zeroth-order infinitesimal of $\delta/R$.

Rotation of MS: Similarly, we consider the problem of an MS rotating without translation ($U = 0$) on a patterned plane, and Equation (1) can be expressed as $V F^r = \mu V^2 U', \nabla \cdot U' = 0$, where the superscript “$r$” represents the rotation (without translation) of the sphere.

Similar to the previous section (Translation of MS) and following the work of Goldman et al.,[21] for an MS rotating on a patterned plane, as shown in Figure 3, the velocity boundary conditions on sphere surface ($S$) and patterned plane ($P$) can be expressed as

$$S \text{(sphere surface)}$$

By solving Equation (1) with boundary condition (11), the dimensionless force can be obtained (see details in the Supporting Information)

$$F^r = -\frac{15}{4} \left(\alpha + \frac{4U_p}{\Omega R}\right) \ln \left(\frac{\delta}{R}\right) + O\left(\frac{\delta}{R}\right)^0$$

where $O(\delta/R)^0$ represents the zeroth-order infinitesimal of $\delta/R$.

Mechanism of Motion Enhancement Due to Microstructures: There are four types of motion behaviors for different microspheres on different planes (i.e., SS moving on smooth plane, MS moving on smooth plane, SS moving on patterned plane, and MS moving on patterned plane). Here, we define $U_{ij}$, $F_{ij}$, and $F_{ij}^s$ as the forward velocity, dimensionless translation force, and dimensionless rotation force in various situations, respectively, where $i, j$ represent the types of the microsphere and plane, respectively. That is, $i, j = s, i, j = m, j = s$, and $j = p$ represent the SS, MS, smooth plane, and patterned plane, respectively. For a given sphere (fixed radius $R$) and a constant magnetic frequency $f_0 (\Omega = 2\pi f_0)$, the ratio of the forward velocity of different types can be obtained by Equation (4) as

$$\frac{U_{ij}}{U_{kj}} = \frac{F_{ij}^s}{F_{kj}^s}$$

We then analyze the four types of motion behaviors based on Equation (10) and (12) to reveal the mechanism of motion enhancement due to microstructures. 1) SS moving on smooth plane: In this situation, $\alpha = \beta = 1$, $U_p = 0$, and Equation (10) and (12) are expressed as $F_{ss}^r = (8/15) \ln(\delta/R) + O(\delta/R)^0$, $F_{ms}^r = (-2/15) \ln(\delta/R) + O(\delta/R)^0$, which are the same results as in the previous study.[21] When an SS swims nearby a smooth plane in the presence of a rotating magnetic field, the relationship between the forward velocity $U_{ss}$ and angular velocity $\Omega$ can be obtained as $U_{ss} = (8/15) \ln(\delta/R) + O(\delta/R)^0$. 2) MS moving on smooth plane: In this situation, $U_p = 0$ while $\alpha, \beta \neq 1$, and Equation (10) and (12) are expressed as $F_{ss}^r = (4/15)(\alpha + \beta) \ln(\delta/R) + O(\delta/R)^0$, $F_{ms}^r = (-2/15) \ln(\delta/R) + O(\delta/R)^0$, $F_{ss}^r = (8/15) \ln(\delta/R) + O(\delta/R)^0$. According to Equation (9) and (13), when moving on a smooth plane, the ratio of the forward velocity of an MS to an SS is

$$\frac{U_{ms}}{U_{ss}} \sim \frac{2\alpha}{\alpha + \beta} > 1$$

Namely, when moving on smooth plane, MS is faster than SS due to the tangential slippage and the interaction between fluid and microstructures, which is consistent with the experimental results in Section 2.2.1.
indicating that the speed of SS on patterned plane is faster than on smooth plane due to the slippage of the surface. 4) MS moving on patterned plane: In this situation, $\alpha, \beta \neq 1$, $U_p > 0$. Equations (10) and (12) are expressed as $F_{m,p}^t = ([4/15] + \beta) - (8/15)\left(\frac{U_p}{U_{m,p}}\right)\ln(\delta/R) + O(\delta/R)^0$, $F_{m,p}^s = -(2/15)(\alpha + 4U_p/\Omega R)\ln(\delta/R) + O(\delta/R)^0$, as $F_{s,p}^t = (8/15)(1 - U_p/U_{s,p})\ln(\delta/R) + O(\delta/R)^0$, $F_{s,p}^s = -(2/15)[1 + 4U_p/\Omega R]\ln(\delta/R) + O(\delta/R)^0$, and $F_{m,s}^t = (4/15)(\alpha + \beta)\ln(\delta/R) + O(\delta/R)^0$, $F_{m,s}^s = (2/15)(\alpha + \beta)\ln(\delta/R) + O(\delta/R)^0$.

Thus, according to Equation (9) and (13), in the case of an MS moving on a patterned plane, the performance of motion enhancement is reflected by two aspects

$$ U_{m,p}^t \sim \frac{1}{\alpha + \beta} - \frac{2\alpha R + 8U_p}{\Omega R + 8U_p} > 1 $$

$$ U_{m,s}^t \sim \frac{1}{\Omega} - \frac{8U_p}{\alpha} > 1 $$

Namely, the forward velocity of MS moving on patterned plane is faster than either SS or on smooth plane due to the slippage on structured surface of sphere or plane, respectively.

Furthermore, based on Equation (15) and (16), it can be demonstrated that the velocity of the MS on the pattern plane is much higher than that of the SS on the smooth plane, that is

$$ \frac{U_{m,p}^t}{U_{s,p}^t} = \frac{U_{m,s}^t}{U_{s,s}^t} > \frac{U_{m,s}^t}{U_{s,p}^t} > 1 $$

which is consistent with the experimental results in Section 2.2.2.

2.4. Determination of Motion Behaviors

2.4.1. Quantification of Parameters $\alpha, \beta$

The introduced parameters $\alpha$ and $\beta$ denote the slippage of fluid on the microstructure surface and the interaction between fluid and microstructures, respectively. They can be estimated from theory and experimental data. For the MS moving on the smooth plane, the normalized scalar torque components are expressed as (see details in the Supporting Information) $T^t = -(1/20)(\alpha + \beta)\ln(\delta/R) + O(\delta/R)^0$ and $T^s = (2/5)\alpha\ln(\delta/R) + O(\delta/R)^0$.

Thus according to Equation (5), the step-out frequency of the SS ($\Omega_s$) and the MS ($\Omega_m$) is given by $\Omega_s \sim V_{smo}[M]|B|/[3\pi R^3\ln(\delta/R)]$ and $\Omega_m \sim V_{str}[M]|B|/[3\pi R^3\ln(\delta/R)]$, where $V_{smo}$ and $V_{str}$ denote the magnetic film volume of SS and MS, respectively. Because the magnetic film is sputtered on the surface of microsphere, and the thickness (150 nm) is quite thin relative to the radius of the microsphere ($40 \mu$m), $V_{smo}$ and $V_{str}$ can be expressed as $V_{smo} = 4\pi R^2_{tm}$ and $V_{str} = 4\pi(R - h)^2 + 4\phi h R^2/d_{tm}$, where $d_{tm} = 150$ nm is the thickness of the Ni magnetic film. Thus, we can get $\Omega_m/\Omega_s \sim [(1 - h/R)^2 + 4\phi h/d/\alpha]$. For an MS with micropillar height $h = 10 \mu$m and solid fraction $\phi = 0.08$, we can obtain $\Omega_m/\Omega_s \sim 1.36/\alpha > 1$ as $\alpha \leq 1$. Namely, the step-out frequency of the MS is higher than the SS. Moreover, the step-out frequencies of MS and SS on smooth plane can be obtained from the experimental data, which are 10 and 6 Hz in our case, as shown in Figure 2c, respectively. Therefore, $\alpha \sim 0.82$. Note that for a given MS with fixed $h$ and $\phi$, $\Omega_m$ depends only on $\alpha$. As $\alpha$ denotes the slippage of fluid over the surface of microsphere, slippage dominates the step-out frequency.

Furthermore, according to Equation (14) and Figure 2c, the linear fittings of forward velocity with respect to the external field frequency below step-out frequency are shown in Figure 4 for both the MS and the SS moving on smooth plane, which presents the fitting slopes are 12.96 for SS and 19.17 for MS, respectively. We can get $U_{ms}/U_{ss} \sim 2/(1 + \beta/\alpha) = 19.17/12.96$; thus, $\beta/\alpha \sim 0.35$ and $\beta \sim 0.29$. According to Equation (8), the slip velocity of fluid in tangential direction is $U_s \sim 0.18 U \sin \phi$ as $\alpha \sim 0.82$. Thus, the effect of fluid slip on tangential velocity is 18%. As $\beta \sim 0.29$, the influence of radial fluid flow in and out of microstructures on radial velocity is 71%. Therefore, the main reason for the enhanced translation velocity lies in the radial flow in and out of the microstructures and the fluid slip in the tangential direction, which reveals the mechanism of motion enhancement due to the surface structures on microsphere.

2.4.2. Influence Factors to Parameters $\alpha, \beta$

To further investigate the influence factors to parameters $\alpha$ and $\beta$, it is necessary to study the motion enhancement of MSs walking on smooth plane under different microstructure sizes and surrounding fluids. Here, the micropillar height $h$, solid fraction $\phi$, and surrounding fluids have been changed.

As $\alpha$ denotes the slippage, it can be regulated by adjusting the slip velocity. Here, we decrease the slippage through increasing the wettability between fluid and MS. To be more specific, isopropanol (IPA) was selected and mixed with DI water to change the wettability. The larger the volume fraction of IPA, the better...
the wettability of the microspheres.\cite{68} Thus, the slippage decreases as the volume fraction of IPA increases. According to Equation (8), the parameter $\alpha$ increases as the volume fraction of IPA increases, which results in the decrease of the step-out frequency. Figure 5a shows the variations of $\alpha$ and $\beta$ as a function of the volume fraction of IPA, and Figure 5b,c shows the effect of volume fraction of IPA on the motion performances of the MS (micropillar height $h = 10\,\mu m$ and solid fraction $\phi = 0.08$) and the SS, respectively. It is experimentally demonstrated that as the volume fraction of IPA increases, $\alpha$ increases and the step-out frequency decreases, which is consistent with our theoretical predictions.

Parameter $\beta$ represents the interaction between fluid and microstructures, which can be tuned by changing the viscosity of surrounding liquid. Increasing the liquid viscosity will decrease the interaction between fluid and MS, which results in the increase in $\beta$ according to Equation (7). Here, glycerol was added into the DI water to increase the viscosity.\cite{69} Figure 5d shows the variations of $\alpha$ and $\beta$ as a function of the volume fraction of glycerol, which experimentally demonstrates that $\beta$ increases as the viscosity increases. However, $\alpha$ increases as the viscosity increases. Thus, both the step-out frequency and the forward velocity decrease as the volume fraction of glycerol increases, as shown in Figure 5e,f.

Figure 5. The influence of the properties of the surrounding liquid on parameters $\alpha$ and $\beta$ and the motion performances of microspheres moving on a smooth plane. a) Variations of $\alpha$ and $\beta$ as a function of the volume fraction of IPA. Forward velocities of b) SSSs and c) MSs as a function of the magnetic field frequency at different volume fractions of IPA. d) Variations of $\alpha$ and $\beta$ as a function of the volume fraction of glycerol. Forward velocities of e) SSSs and f) MSs as a function of the magnetic field frequency at different volume fractions of glycerol.
The size of the microstructure on the sphere will also affect the parameters $\alpha$ and $\beta$. First, experiments for three heights of micropillar $h = 5$, 10, and $15\ \mu m$ were performed with the same solid fraction $\phi = 0.08$. When the MSs move in DI water under the magnetic field strength of 14 mT, the variations of $\alpha$ and $\beta$ as a function of $h$ are shown in Figure 6a, and the effect of $h$ on the motion performance is shown in Figure 6b. For one thing, the increase in $h$ results in the decrease in $\alpha$ and the increase in the step-out frequency $\Omega_m$, which is consistent with the theoretical prediction that the smaller the $\alpha$, the better the motion performance (larger $\Omega_m$). For the other thing, the forward velocity of the microsphere in the case of $h = 10\ \mu m$ is always greater than the other two cases when $\Omega < \Omega_{\text{step-out}}$, which can be well explained by Equation (14) that the smallest $\beta$ in the case of $h = 10\ \mu m$ indicates the highest velocity. Note that the increase in $h$ will increase the magnetic film volume of MS ($V_{\text{str}}$), which also contributes to the motion enhancement. Second, the experiments for three solid fractions of micropillar $\phi = 0.04$, 0.08, and 0.15 were performed with the same micropillar height $h = 10\ \mu m$. Figure 6c shows the variations of $\alpha$ and $\beta$ as a function of the solid fraction $\phi$. As $\phi$ increases, $\alpha$ and $\beta$ increase, indicating both the slip velocity on the microstructure surface and the interaction between fluid and microstructures decrease. The effect of $\phi$ on the motion performance is shown in Figure 6d. It is noted that the step-out frequency increases due to the increase in magnetic film volume rather than the change of $\alpha$, whereas the forward velocity decreases due to the increase in $\beta$ according to Equation (14).

The above-mentioned experimental results show that the parameters $\alpha$ and $\beta$ can exhibit different values for different microstructure sizes and fluids and, thus, can be tuned to obtain different motion performance.

2.4.3. Effect of Parameters $\alpha$, $\beta$ on Motion Behaviors

Apart from the motion enhancement discussed earlier, the variations of $\alpha$ and $\beta$ may result in other motion behaviors of surface walkers. We first discuss the effect of $\alpha$ based on Equation (8) to the slip velocity on MS surface $U_s$ as the following: 1) $\alpha = 1$ represents the case without slippage; thus, $U_s = 0$. This situation corresponds to the smooth microsphere moving nearby a plane. 2) $\alpha < 1$ represents the case with slip velocity $U_s > 0$; that is, the slip velocity of the fluid on MS surface is in the same direction as the sphere velocity. This situation is the same as our experiment that an MS moving nearby a plane in still fluid. In this circumstance, the step-out frequency of MS is greater than that of the SS. 3) $\alpha > 1$ represents the case with slip velocity $U_s < 0$. This

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Figure 6. The influence of the microstructure size on the parameters $\alpha$, $\beta$ and the motion performance of MSs moving on a smooth plane. a) Variations of $\alpha$ and $\beta$ as a function of the micropillar height $h$. The insets are the SEM images of the MSs with $h = 5\ \mu m$, 10, and $15\ \mu m$, respectively. b) Forward velocity with respect to the magnetic field frequency for different micropillar heights. c) Variations of $\alpha$ and $\beta$ as a function of the solid fraction $\phi$. The insets are the SEM images of the MSs with $\phi = 0.04$, 0.08, and 0.15, respectively. d) Forward velocity with respect to the magnetic field frequency for different solid fractions.
situation corresponds to that the slip velocity of the fluid on MS surface is in the opposite direction as the sphere velocity. In this circumstance, the step-out frequency of MS may be less than that of the SS.

Meanwhile, according to Equation (7), $\beta = 1$ represents the SS with no radial flow in and out of sphere. The velocity component of the flow field around the sphere in the $y$-direction is expressed as $u_y = (\beta - \alpha) U \cos \phi \sin \phi$. We then discuss the effect of $\beta$ as the following: 1) $\beta < \alpha$ represents that the flow velocity around the microsphere on $x$-$y$ plane is in the same direction as the forward velocity of sphere, as shown in Figure 3d. In this circumstance, the translation velocity of MS is greater than that of SS, indicating that the introduction of microstructures contributes to the enhancement of velocity of the MS. 2) $\beta > \alpha$ represents that the flow velocity around the microsphere on $x$-$y$ plane is in the opposite direction to the forward velocity of sphere. For instance, there is a flow with a relatively high speed in the opposite direction to the movement of sphere, or there is a backflow around the sphere. In this circumstance, the translation velocity of MS is less than that of SS, indicating that the introduction of microstructures will reduce the velocity of the MS.

In our experiments, the fluid is still, and thus, the projection of streamlines around the MS surface on $x$-$y$ plane is the same as that shown in Figure 3d, i.e., $\beta < \alpha$, revealing an enhancement of motion for the microsphere due to the introduction of surface microstructures.

3. Conclusion

The current work explores how surface microstructures affect the motion behavior of micro-objects. A microsphere with micropillars array patterned on its surface has been introduced and driven by a rotating magnetic field on a smooth plane and a patterned plane, respectively, in which the translation velocity has been recorded. There is almost a 2.2 and 4.1 times increase in the forward velocity for the MS walking on the smooth plane and the patterned plane compared with the SS walking on the smooth plane, respectively. Through theoretical analyses, the mechanism for the enhanced translation velocity is revealed that the motion enhancement is attributed to the slippage of fluid on the microstructured surface and the interaction between fluid and microstructures due to fluid flowing in and out of the microstructures. Furthermore, two parameters $\alpha$ and $\beta$ have been introduced to denote the effects of the slippage and the interaction between fluid and microstructures, respectively. The parameters can be estimated from theory and experimental data. Furthermore, the values of $\alpha$ and $\beta$ can be tuned by designing the microstructures and the surrounding liquid, so as to controllably adjust the motion performance of the MSs. The current analyses on the motion enhancement due to the introduction of surface microstructures can be extended to the surface walkers with other shapes, and the result of enhanced translation velocity due to microstructures can be used for biomedical applications, such as faster biomimetic microdevices, targeted drug delivery, and cell manipulation in complex aqueous environments.

4. Experimental Section

Fabrication of Microspheres: The microspheres were fabricated with SU-8 photoresist on glass substrate by means of 3D DLW (Photonic Professional GT, Nanoscribe GmbH). The fabrication of the sample required several steps. First, the glass substrate was cleaned with isopropyl alcohol (IPA) by ultrasonic, and then rinsed with DI water. Then, the SU-8 photoresist was spin-coated on the glass substrate with a thickness of 100 $\mu$m at a speed of 1000 rpm. Afterward, the sample was pre-baked with a hot plate at 65 °C for 10 min followed by a second step at 95 °C for 30 min, and then loaded in the laser lithography system (Photonic Professional GT, Nanoscribe GmbH) and exposed to laser beam with a center wavelength of 780 nm. The Galvo (layer by layer) Scanning Writing Mode was used to fabricate the sample with high efficiency and precision. After the lithography process, the sample was post-baked with a hot plate at 65 °C for 1 min followed by a second step at 95 °C for 10 min and developed for 12 min in SU-8 Developer and rinsed with IPA and DI water. Finally, the sample was coated with a 150 nm thickness Ni film by direct current (DC) sputtering (Q150T S, Quorum Technologies Ltd., UK). During the coating process, the sample stage was tilted to an angle, which allows the bottom half of the microsphere to be sputtered with magnetic materials. Meanwhile, the sample stage can perform a rotational motion around the vertical direction as well as the direction perpendicular to the inclined plane with an angular velocity, which ensures a relative uniformity of sputtering material on the microsphere. In our experiments, the sample stage rotated with a rotational speed of 70 rpm and a tilt of 30° to achieve a relatively uniform Ni coating over entire surface of the microspheres as much as possible. However, a coating with non-uniform thickness distribution existed anyway; i.e., the Ni film was thicker on the top than the bottom of the microsphere (see Supporting Information). The same sputtering process was carefully implemented for coating Ni films on all the microspheres to ensure that the thickness distributions of Ni film from top to bottom are the same for both MS and SS. It can be demonstrated that no matter the Ni coating is uniform or not, the conclusion on the motion enhancement due to the microstructures is valid (see details in the Supporting Information).

Fabrication of Patterned Plane: The patterned plane was fabricated using the lithography and silicon etching technology on the Si substrate. First, the Si substrate was cleaned with acetone and IPA by ultrasonic for 5 min and then rinsed with DI water. Second, the photoresist RZJ-304 was spin-coated on the Si substrate at a speed of 4000 rpm for 30 s. After a pre-bake process (90 s at 100 °C), the sample was exposed to UV light with a center wavelength of 365 nm using a laser lithography system (MJ84, SUSS MicroTec, Germany). Third, the sample was developed for 30 s in RZX-3038 and post-baked 120 s at 100 °C. After this, the patterned Si substrate was etched with the silicon etching technology, and then, the remaining photoresist was cleaned with acetone by ultrasonic.

Microspheres Manipulation in Magnetic Actuation System: The microspheres were manipulated in a magnetic actuation system (MFG-100, MagneNetBox AG, Switzerland). The microspheres and patterned Si plane are immersed in DI water inside a plastic reservoir (30 × 16 × 5 mm). The reservoir is placed at the center of the magnetic actuation system to ensure the uniformity of the magnetic field. Before each experiment, a microprobe (T-4-22, GGB Industries, Inc.) with a sharp tip was used to detach and transport microspheres from the glass substrate onto the Si substrate. MSs and SSs were used for characterizing the swimming properties on smooth plane and patterned plane. Each microsphere was tested at least three times. The forward/rotational velocity and step-out frequency were measured.

Supporting Information

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

This work was supported by the Beijing Natural Science Foundation under grant no. L172002 and National Natural Science Foundation of China (NSFC) under grants nos. 9184201, 11521202, 11988308, 11872004, and 11802004.

Conflict of Interest

The authors declare no conflict of interest.

Keywords

microrobots, microspheres, microstructured surfaces, motion enhancement, surface walkers

Received: October 7, 2020
Revised: December 14, 2020
Published online: March 18, 2021
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