Rational Design of Reversible Redox Shuttle for Highly Efficient Light-Driven Microswimmer

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ABSTRACT: The light-driven micro/nanomotor (LMNM) is machinery that harvests photon energy and generates self-propulsion in varieties of liquid media. Though visions are made that these tiny swimming machines can serve future medicine for accurate drug delivery and noninvasive microsurgery, their biomedical application is still impeded by the insufficient propulsion efficiency. Here we provide a holistic model of LMNM by considering (i) photovoltaic, (ii) electrochemical, and (iii) electrokinetic processes therein. Such a quantitative model revealed the pivotal role of reaction kinetics and diffusion properties of shuttle ions in the propulsion efficiency of LMNM. With the guidance of this model, a group of ferrocene-based reversible redox shuttles, which generate slow-diffusion ions, was identified, showcasing a high locomotion velocity of ∼500 μm/s (∼100 body length per second) at an ultralow concentration (70 μM). Owing to the in-depth understanding of the fundamental energy conversion processes in LMNM, we anticipate that the development of other high-performance supporting chemicals and LMNM systems will be greatly motivated, foreseeing the advent of LMNM systems with superior efficiency.

KEYWORDS: silicon nanowire, light-driven microswimmer, redox shuttles, efficiency, biocompatibility

Chemomechanical actuation by ionic streams is ubiquitously identified in natural microbes. Protons (H+) inherent in aqueous media are utilized by many microorganisms such as Streptococcus to generate a proton-motive force and twiddle their flagella.1 The nonproton ions, such as Li+ and Na+, are also found responsible for the rotation of biotic motors in the marine bacteria such as Vibrio alginolyticus.2 As the artificial counterparts, proton and heavy metal ions, like Cu2+, can drive abiotic micromachines.3 Such chemically powered artificial micro/nanoswimmers (MNNs), being capable of converting chemical energy into mechanical motion, are considered as promising candidates for accurate and noninvasive medicine.4–8 To that end, several prerequisites, particularly the high propulsion efficiency of supporting chemicals,9,10 should be met. In the past decades, researchers in this area endeavored to achieve this target. To provide strong mechanical thrust, high-energy rocket fuels such as hydrogen peroxide (H2O2)11–14 and hydrazine (N2H4)15–17 have been extensively exploited, which can vigorously react and propel the MNM. On the other hand, MNMs based on fuels naturally present in biofluids, such as H2O, glucose, and urea, were developed by designing the enzymatic or photocatalytic propulsion.18–23 For practical usage of MNMs, further advancement of fuel efficiency is highly desirable,24 which may also mitigate the fuel toxicity issue by lowering the dose. In this respect, some theoretical study has been reported to improve the performance of catalytic motors,25 several highly efficient bubble-free bimetallic nanomotors with enhanced efficiency in low concentration halogen were also exploited.3,26 However, few studies provided experimentally corroborated guidelines for supporting chemicals to improve the efficiency of MNMs.

As a thermodynamic driving force, light has been explored to drive MNMs.27–30 To date, light-driven micro/nanomotors (LMNNs) have been demonstrated based on different...
propulsion mechanisms, involving bubble propulsion, thermal propulsion, diffusiophoresis, and electrophoresis driven, etc.\textsuperscript{31−36} Compared to other MNMs, LMNMs promise additional degrees of freedom due to light’s variable intensity, frequency, polarization, and propagation direction with spatiotemporal accuracy, which enables excellent controllability and programmability of LMNMs.\textsuperscript{34,35,37−40} In particular, LMNMs offered a potential solution to the efficiency-dose dilemma by decoupling the energy source from the supporting chemicals. Specifically, the propulsion is powered by the incident light instead of the consumption of fuels, while the supporting chemical merely serves as the redox shuttle to support the surface redox reaction on LMNM, without net consumption.\textsuperscript{39} In principle, high propulsion thrust can be provided under ultralow chemical dose, providing desirable biocompatibility. However, this highly efficient redox shuttle has hitherto not been reported, while H\textsubscript{2}O\textsubscript{2} is still the chemical of choice in most LMNM studies,\textsuperscript{41−43} and the detailed understanding of the energy cascade in LMNM systems is highly desirable.

To meet the prerequisites of biocompatible LMNMs, three criteria must be taken into consideration: (1) moderate light intensity to avoid heat damage to living tissues in view of the limited light penetration into the tissue, (2) low concentration of supporting chemical to ensure minimal toxicity, and (3) high photomechanical energy conversion efficiency to achieve enough propulsion thrust. In this study, we present a quantitative model for LMNM by combining the photoelectrochemical and electrochemo-mechanical energy conversion processes therein. With the help of our model, the kinetic constant of electrochemical reaction and the diffusion coefficient of the charged ionic shuttle are identified as the determining factors for the improvement of propulsion efficiency. Based on the finding, a class of ferrocene-based (Fc) reversible shuttles were demonstrated with advanced propulsion efficiency and low cytotoxicity.

**RESULTS AND DISCUSSION**

**Photo to Electrochemical Model of LMNM.** The energy conversion in LMNM system can be generalized into three steps as illustrated in Figure 1: (i) photon absorption and photovoltage generation through the photovoltaic effect, (ii) the electrochemical reaction driven by the photovoltage and the local buildup of charged ionic shuttles, and (iii) the establishment of electric field by charged shuttles and the propulsion of the LMNM by electrokinetic force. Inset: the representative ionic shuttles for LMNMs.

Figure 1. Schematic of the light-driven microswimmer that converts incident photon into mechanical propulsion with the aid of redox shuttles. The energy cascade in the core−shell p/n junction silicon nanowire (Figure S1) can be regarded as three steps: (i) the capture of incident photons and photovoltage generation, (ii) the electrochemical reaction driven by the photovoltage and the local buildup of charged ionic shuttles, and (iii) the establishment of electric field by charged shuttles and the propulsion of the LMNM by electrokinetic force. Inset: the representative ionic shuttles for LMNMs.
light intensity $I$ as follows (see Supplementary Discussion for details):

$$\Phi I \approx j \left( \frac{J}{2J_0^c} + \left( \frac{J}{2J_0^a} \right)^2 + 1 \right) \left( \frac{mJ}{2J_0^c} + \left( \frac{mJ}{2J_0^a} \right)^2 + 1 \right)^{-1/2} \left( \frac{j_{da}j_{dc}}{(j_{dc} - J)(j_{da} - J)} \right)^{1/2} + J \right)$$

(1)

where $\Phi$ and $n$ are the external quantum efficiency and the ideality factor of the photovoltaics respectively; $m$ is the area ratio of the cathode to anode electrode; $J_0^c$ and $J_0^a$ are the cathode and anode exchange current density, respectively; $j_{da}$ and $j_{dc}$ are the cathode and anode diffusion-limited current density, respectively.

In eq 1, the exchange current density $j^0$ is further determined by the concentration and the kinetic constant of the redox chemical: $j^0 = FkC_0C_R^\alpha$ while the diffusion-limited current density $j_d$ is determined by the diffusion coefficient of redox chemical: $j_d = zFDi_0 \frac{c_i}{d}$, where $F$ is the Faraday constant, $k$ is kinetic constant, $\alpha$ and $\beta$ are transfer coefficients and can be assumed as 0.5 for a reversible redox couple, $C_0$ and $C_R$ are the oxidant and reductant concentrations, respectively; $z$ is the number of electrons in the reaction, $D_i$ is the diffusion coefficient of the reactant $i$, $c_i^0$ is the bulk concentration of the reactant $i$, and $d$ is the thickness of the diffusion layer.

Since the current density $J$ indicates the driving power for a microswimmer, the LMNM speed increases linearly at low illumination conditions and saturates at a high light intensity, as depicted by eq 1, with contributions from both concentration and the kinetic constant (see Supplementary Discussion). To facilitate the understanding of these effects, the photoresponse of photoelectrochemical current density can be visualized by a free interactive online tool (https://www.desmos.com/calculator/pmdfz5pp2l, see Supplementary Discussion). Overall, the determining factor is the kinetic constant $k$ (Supplementary Discussion), which can greatly improve the current density and facilitate operation under low light intensity as well as low supporting chemical concentration.

Large Kinetic Constant for Efficiency Improvement.

With the quantitative model, the microswimmer speed can be predicted by eq 1, which enables the comparison and optimization of supporting chemicals. The diffusion coefficients and kinetic constants of five different redox chemicals used in this study are given in Table S1, which were measured...
Figure 3. Motility of the same microswimmer in supporting chemicals shows much lower concentration needed for MFc. (a–c) The migration trajectory with different redox shuttles (70 μM MFc, 30/3 mM H₂Q/BQ, and 30 mM H₂O₂) by stacking sequential frames within 1s (Movie S1). The solid dots denote the head positions of the microswimmer in each frame. The arrows indicate the motion direction. The scale bar is 10 μm. (d) Migration speed of the same microswimmer under varies of chemical concentration with constant light intensity (∼420 mW/cm²). The theoretical curves are fitted from eq 1 with the corresponding photovoltaic and electrochemical parameters in Table S1. The H₂O₂ fitting curve was corrected based on the real kinetics constant under different concentrations (Figure S13). Error bars represent the standard deviation of the speed. (e) The intersection points of the photovoltaic (black) and electrochemical curves (red, purple, and blue correspond to 70 μM MFc, 30/3 mM H₂Q/BQ and 30 mM H₂O₂, respectively) highlighted low overall current density in MFc.

electrochemically by rotating-disk-electrode (RDE) voltammetry (Supplementary Methods, Figures S3–S10). To demonstrate the universal validity of this model, we first selected multiple microswimmers with a noticeable difference in performance and tested their light response. As shown in Figure 2a–c, despite the variation among samples, the normalized speed versus light intensity relationships were obtained with different redox chemicals, which matched well with the J–V curves (solid lines) given by eq 1. Further, the same microswimmer migration behavior was tested with different redox chemical concentration and light intensity (Figure 2d–f), where the measured speed is well-predicted by eq 1. The relatively large deviation of H₂O₂ can be attributed to the kinetic constant variation under high concentration (Figure S12). Nevertheless, it is notable that for all redox shuttles, the swimmer migration speed increases linearly at a low light intensity and saturates at high intensity, which aligns with the model prediction. Moreover, due to the much higher kinetic constant (k₀ and k₉) (Table S1), the ferrocenemethanol (MFc) provides the maximum speed at a much lower light intensity, which is desirable considering the moderate light intensity criterion.

Since both the kinetic constant of the redox reaction and the diffusion coefficient of the charged ionic shuttle will influence the current density J and microswimmer motility, it is valuable to clarify the effect of kinetic constant without interference from the latter. Therefore, we plotted the current density J with k₀ and k₉ as variables, keeping all the other parameters constant, including the diffusion coefficient (Figure 2g). It is evident that the raised k₀ and k₉ will increase J, thus promoting the flux generation of ionic shuttles and the driving force. As an experimental proof, the same silicon microswimmer was transferred and tested in MFc and OH-TEMPO due to their distinct kinetics constant and similar diffusion coefficient of the charged shuttles (see Figure S4 and Table S1). It can be seen that the swimmer speed of the more kinetics-active MFc greatly surpassed that of OH-TEMPO even with hundreds of times lower dose, particularly under low light intensity conditions (Figure 2h), confirming the high reaction kinetics of MFc and its capability to effectively propel the LMNM under moderate light intensity.

Sluggish Diffusion for Efficiency Improvement. Based on criterion 2, a lower supporting chemical concentration is preferred for the LMNM operation, thus the chemical concentration dependence of the same microswimmer was tested in MFc, H₂Q/BQ, and H₂O₂ solution. It can be clearly noted that the MFc can support comparable microswimmer migration with over 400 times lower concentration than H₂Q/BQ and H₂O₂ (Figure 3a–d). However, although MFc exhibits much higher kinetic constant, the predicted current density in MFc (70 μM) is still ∼30 times lower than that in H₂Q/BQ (30 mM) or H₂O₂ (30 mM), as shown in Figure 3e, implying other factors, in addition to the kinetic constant, for the enhancement of propulsion efficiency in MFc.

In the self-electrophoretic microswimmer, the chemical reaction provided the unbalanced ion distribution around the microswimmer, and the charged ions conditioned the electric field applied to the microswimmer, which further regulated the locomotion through an electrokinetic force. Once the ion flow is generated, it will be transported into the surrounding media by convection, diffusion, and electrophoresis migration.24,44 Without the external flow field, the self-generated electroosmosis flow (EOF) is the only source contributing to the convection around a swimmer. The swimmer is further propelled by pumping the surrounding fluid backward according to Newton’s third law. Thus, only the convection contributes to propulsion, while the diffusion and electrophoresis migration of the charged ions just diminish the ions gradient and cause the loss of energy.
Quantitatively, the relationship between the swimmer migration speed \( U \) and the diffusion coefficient of the charged ionic shuttle \( D_{\text{ion}} \) can be given as follows (see Supplementary Discussion for detail):

\[
U \propto \frac{J}{D_{\text{ion}}}
\]

This equation reveals that the migration speed is inversely proportional to the diffusion constant of the ionic shuttle, which aligns with the reported results\(^2\)\(^4\)\(^4\)\(^5\) and can be used to predict the LMNM propulsion speed (visualization by a free interactive online tool, see Supplementary Discussion). Considering the diffusion coefficient of MFc\(^+\) is only \( \sim 1/37 \) of that of H\(^+\), the lower current density in MFc can be well compensated, leading to the overall enhancement of per-reaction efficiency. More importantly, this result suggested that a sluggish diffusion coefficient of the charged ionic shuttle is favorable for high-speed microswimmer propulsion.

To have an in-depth understanding of the \( D_{\text{ion}} \), we numerically investigated our microswimmer system using COMSOL Multiphysics. For high diffusion coefficient ionic shuttle such as H\(^+\), only 0.07% of the ion flux is directed toward the convection (Figure 4a), which could be a major factor for the low-efficiency propulsion of the LMNMs fueled by H\(_2\)O\(_2\) and N\(_2\)H\(_4\). In contrast, MFc\(^+\) with sluggish diffusion enables 26.8% of ion flux toward convection, boosting per-reaction propulsion efficiency by \( \sim 380 \) times (Figure 4a). This enhancement can also be visualized by comparing the electric field around the microswimmer in MFc (Figure 4b) and H\(_2\)O\(_2\)/BQ (Figure 4c) with the same total ion flux, where a stronger electric field can be established by the MFc\(^+\) shuttle.

In light of the above results, we envisage that the ionic product with more sluggish diffusion can further increase the propulsion efficiency of microswimmer. Accordingly, ferrocenylmethyl β-D-glucopyranoside (GFc) is synthesized as a comparison to MFc (synthesis process, see Supplementary Methods). Due to its larger molecular hydrodynamic radius, a smaller \( D_{\text{ion}} \) is expected, hence, a higher microswimmer efficiency. As shown in Figure 4d, the diffusion coefficient of GFc\(^+\) is \( 2.00 \pm 0.017 \times 10^{-6} \) cm\(^2\)/s, which is smaller than MFc\(^+\) (2.51 \( \pm \) 0.005 \( \times \) 10\(^{-6} \) cm\(^2\)/s). As expected, the improved per-reaction propulsion efficiency is observed for GFc over MFc (Figure 4e). The locomotion results of MFc and GFc successfully demonstrated the feasibility of using a kinetics-active and diffusion-sluggish redox shuttle to support the LMNM propulsion under ultralow concentration and subdued toxicity. By using well-developed redox-active derivatives such as dendrimers and assemblies with an even larger hydrodynamic radius,\(^4\)\(^6\) even higher propulsion efficiency could be expected.
Fc-based redox shuttles. Specifically, the MFc shows outstanding efficiency over that of H$_2$Q/BQ and H$_2$O$_2$ with much lower concentration (Figure 5a). For clarity, the highest photomechanical efficiency of the three redox shuttles is presented in Figure 5b, where the experimental propulsion efficiency in MFc is about 10 times higher than in H$_2$Q/BQ and H$_2$O$_2$. In comparison with the numerical result (∼300 times increase in Figure 4a), this lower efficiency enhancement can be attributed to the smaller current density due to the low concentration of MFc. With our silicon microswimmer, speed over 500 μm/s (∼100 body length per second) was observed in 70 μM MFc (inset image of Figure 5b, Movie S2), which is the record of self-electrophoretic LMNM and the corresponding efficiency is about 2.3 × 10$^{-6}$. Although this efficiency is still low, 3 orders of magnitude enhancement have been achieved in comparison with the efficiency (usually on the order of 10$^{-10}$–10$^{-9}$) of bimetallic catalytic motors. With optimized catalyst and bandgap engineering for photoelectrochemical process, further improvement of efficiency can be expected.

Compared with the enzymatic motors powered by naturally available chemicals in the human body, the biocompatibility of noninherent chemicals through injection or oral intake is of great concern. The cytotoxicity of MFc, H$_2$Q/BQ, and H$_2$O$_2$ were compared using human U-2 OS cell line with standard IC$_{50}$ (half-maximal inhibitory concentration) measurement (see Supplementary Methods for details and Figure S15). MFc has been commonly used as electrochemical mediators to study the cellular toxicity of chemicals in scanning electrochemical microscopy. Due to its low concentration (mM), MFc has no apparent effect on cell viability. With the typical dose used in this study (∼0.1 mM), MFc displayed excellent biocompatibility as shown in the 24-h PrestoBlue Cell Viability assay test (Figure 5c). In contrast, H$_2$O$_2$ (0.01 wt %) and H$_2$Q/BQ (1/0.1 mM) shows toxicity at even lower dose than that required for microswimmer operation. This result, however, will not rule out the potential use of H$_2$Q/BQ or H$_2$O$_2$ for biological applications, which may require further investigation due to the potentially significant tolerance variation between different cell lines. Since both IC$_{50}$ and propulsion efficiency depends on the redox shuttle concentration, the IC$_{50}$ can be plotted against the relative propulsion efficiency to highlight the supporting chemical’s toxicity-efficiency performance. As shown in Figure 5d, MFc shows outstanding propulsion efficiency and appealing relative viability, indicating that the proposed ferrocene-based derivatives can serve as a promising fuel in both energy conversion efficiency and biocompatibility, which foresees the advent of other high-performance supporting chemicals over the toxicity-efficiency dilemma.

**CONCLUSION**

In summary, we proposed a holistic model for LMNN, which can quantitatively predict the performance of supporting chemicals with measurable electrochemistry parameters. The kinetic constant and the diffusion coefficient have been
identified as the key factors to determine the propulsion efficiency of LMNM. Based on this model, a group of redox-active but diffusion-sluggish ferrocene derivatives have been developed for microswimmer, enabling a record speed of ~500 μm/s (~100 body length per second) for self-electrophoretic LMNM at a biocompatible fuel concentration as low as 70 μM. Impressively, the propulsion efficiency of LMNM with the MFC can reach as high as 10−6, which is three orders magnitude higher than that of bimetallic catalytic motors. It can be expected that even higher efficiency is feasible with carefully designed catalytic systems and optimized diffusion properties, providing another paradigm for solving the toxicity-efficiency dilemma. Besides the supporting chemicals, it is noteworthy that ion-tolerance is another critical problem for LMNM operation in high ionic strength body fluids. Recently, we demonstrated that the surface modification with polyelectrolyte could be a route toward enhanced ion tolerance for electrokinetic locomotion.19 We believe, with the continued progress in efficiency and biocompatibility, practical biological and medical applications of LMNMs can be envisioned.

METHODS

Silicon Wire Microswimmer Fabrication, Migration Measurement, and Data Analysis. The silicon wire microswimmer was fabricated by previously reported methods.19 Typically, the silicon nanowire array can be grown by the vapor–liquid–solid (VLS) process. Subsequently, the thermal diffusion method was used to generate the n−Si shell and form the p/n junction (see Supplementary Methods for details). Then, 0.5 nm platinum nanoparticles were deposited on the surface of silicon nanowires as an electrolytically driven on a customized MATLAB program was used for migration speed measurement. Microswimmer speed dependence on light intensity, as the illumination source in all experiments. The intensity of the light was the number of transferred electrons each motion.

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Electrochemistry Characterization. Rotating disk electrode (RDE) voltammetry was used to obtain the diffusion coefficient of fuels. The measurement was conducted on a CHI660E electrochemical workstation (CH Instruments, Inc.) using a standard three-electrode setup with a standard Pt disk working electrode (5 mm diameter), an Ag/AgCl reference electrode, and a Pt wire counter electrode. The RDE test was done in 0.1 M NaCl electrolyte solution with supporting chemical at a suitable scanning range vs Ag/AgCl and a scanning rate of 0.01 V/s. Voltammograms data were analyzed via a Levich plot and yielded the corresponding diffusion coefficient D by the Levich equation:

\[ D = \frac{0.62 n F A t D^{1/3}}{5 \eta (1/n)^{1/2} v} \]  

where \( D \) is the diffusion coefficient of fuel, \( n \) is the number of transferred electrons per redox reaction, \( F \) is Faraday’s constant, \( A \) is the area of the electrode, \( \eta \) is the viscosity of solution, \( v \) is the rotation speed, and \( t_d \) is the bulk concentration of fuels.

To mimic the real conditions in silicon LMNM, the reduction and oxidation kinetic constants were measured using n+ silicon wafer loaded Pt particles as the cathode and p+ silicon wafer as the anode, respectively. For the reduction of ferrocenium derivatives, benzoquinone, and \( \text{H}_2\text{O}_2 \), the RDE voltammograms were measured using a 1 cm² (1 cm x 1 cm) n+ silicon wafer loaded with Pt particles, which was intimately fixed on the surface of Pt disk electrode. Application of the Koutecký–Levich equation:

\[ \frac{1}{i} = \frac{1}{i_0} + \frac{1}{i_{lim}} \]  

which is three orders magnitude higher than that of bimetallic catalytic motors. It can be expected that even higher efficiency is feasible with carefully designed catalytic systems and optimized diffusion properties, providing another paradigm for solving the toxicity-efficiency dilemma. Besides the supporting chemicals, it is noteworthy that ion-tolerance is another critical problem for LMNM operation in high ionic strength body fluids. Recently, we demonstrated that the surface modification with polyelectrolyte could be a route toward enhanced ion tolerance for electrokinetic locomotion.20 We believe, with the continued progress in efficiency and biocompatibility, practical biological and medical applications of LMNMs can be envisioned.

Numerical Simulation of Ion Distribution and Electric Field. The charged ion distribution and fluid speed were simulated using the commercial COMSOL Multiphysics package. For \( \text{H}_2\text{O}_2 \) and hydroquinone/benzoquinone (H2Q/BQ) systems, protons as cations (H+) are generated at p-Si (anode) and consumed at n-Si (cathode) surfaces, respectively. For ferrocene derivatives, the corresponding ferrocenium cations are generated at p-Si (anode) and reduced at n-Si (cathode) surfaces, respectively. The distribution of charged ions is governed by eq S4, which will facilitate the establishment of an electric field and propel the motor motion:

\[ -\varepsilon_r \nabla \phi = \rho_s = F z e_n \]  

where \( \phi \) is the electric potential, \( \epsilon_r \) is the vacuum permittivity and \( e_n \) is the relative electric permittivity of water, \( z = +1, \rho_s \) is the volumetric charge density, \( F \) is the Faraday constant, and \( z_e \) is the concentration of the corresponding cations, respectively. The boundary condition was adopted according to the zeta potential of the Si nanowire (Si surface ~20 mV).21 The steady ionic fluxes on both the anode and cathode surfaces are adopted based on the measured photocurrent density. All the adopted diffusion coefficient constants of related cations are listed in Table S1.

Syntheses and Cellular Viability Assays. The detailed synthesis of OH-TEMPOBF4, ferrocenethyl \( \beta \)-fluoropyranoside, and ferrocenium derivatives, as well as cellular viability assays, are provided in the Supplementary Methods.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsnano.9b08799. Detailed experimental procedures for microswimmer fabrication, chemical synthesis, cellular viability measurement, electrochemistry characterization, microswimmer migration measurement, and numerical simulation; relevant equation discussion for equation deduction process, electrokinetic effect, holistic analysis of speed-determining parameters, and analysis of energy conversion efficiency; additional figures including SEM image, electrochemistry measurement, numerical simulation, microswimmer speed dependence on light intensity are provided.
intensity, cytotoxicity result, ICP-MS curve, UV−vis measurement; and table of electrochemistry parameters (PDF)

Movie S1, LMNM motion in different fuels (MP4)

Movie S2, record of LMNM motion (MP4)

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Notes

The authors declare no competing financial interest.

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