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A catalytic oligomeric motor that walks along a filament track

Mu-Jie Huang^{a)} and Raymond Kapral^{b)}

Chemical Physics Theory Group, Department of Chemistry, University of Toronto, Toronto, Ontario M5S 3H6, Canada

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Most biological motors in the cell execute chemically powered conformational changes as they walk on biopolymer filaments in order to carry out directed transport functions. Synthetic motors that operate in a similar manner are being studied since they have the potential to perform similar tasks in a variety of applications. In this paper, a synthetic nanomotor that moves along a filament track, without invoking motor conformational changes, is constructed and its properties are studied in detail. The motor is an oligomer comprising three linked beads with specific binding properties. The filament track is a stiff polymer chain, also described by a linear chain of linked coarse-grained molecular groups modeled as beads. Reactions on the filament that are catalyzed by a motor bead and use fuel in the environment, in conjunction within the binding affinities of the motor beads to the filament beads, lead to directed motion. The system operates out of equilibrium due to the state of the filament and supply of fuel. The motor, filament, and surrounding medium are all described at microscopic level that permits a full analysis of the motor motion. A stochastic model that captures the main trends seen in the simulations is also presented. The results of this study point to some of the key features that could be used to construct nanomotors that undergo biased walks powered by chemical reactions on filaments. © 2015 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4922926]

I. INTRODUCTION

Directed transport over long distances plays an important role in the biochemistry of the cell. DNA and RNA polymerases translocate along DNA to carry out replication and transcription; kinesin and myosin molecular motors move unidirectionally along tubulin and actin filaments, respectively, transporting cargo chromosomes and vesicles to their destinations. These and other motor proteins are often powered by binding of adenosine triphosphate (ATP) molecules and convert chemical energy into conformational motions that give rise to directed motion along biopolymers or biopolymer filaments.^{1,2} There is a large literature that deals with Brownian ratchet^{3–5} and other models designed to elucidate mechanisms for the directed motion of such motors.^{6–17}

Motivated by such naturally occurring molecular motors, a considerable effort has been made to design controllable synthetic molecular and nanoscale motors that are able to execute directed motion on substrates. Research on this topic can lead to potential applications in nanotechnology, for instance, those involving transport of material or information on the nanoscale along specified paths to specific destinations.¹⁸ Examples of such motors include those that execute directed Brownian motion due to a substrate friction gradient,¹⁹ as well as those that move as a result of ligand-coordinated^{20–22} and light-powered^{23–26} biased diffusion.

A class of biased-diffusive molecular motors is based on the "burnt-bridge" mechanism.^{27,28} In this mechanism, the motor jumps between two nodes on a one-dimensional

track, and the link bridging two nodes may be "burnt" after each jump event. To bias the motor motion, a specific form of the link-burning event is assumed; that is, a link-burning event may occur only when the motor jumps to the node on its right. Once the link is burnt, it cannot be crossed again and the motor will undergo biased diffusion with subsequent jumps to the region without burnt links. Applications of the burnt-bridge model have been proposed. For instance, the biased diffusion of the activated collagenase, matrix metalloprotease (MMP), was found to be in agreement with the results of Monte Carlo simulations of a burnt bridge Brownian ratchet model.²⁹ In addition, DNA molecules that have been engineered to self-assemble with target substrates have been adopted as candidates for specifically designed synthetic nanomotors.³⁰⁻³⁴ DNA machines, such as molecular spiders³⁵ and a self-transported nanoparticle³⁶ that use the properties of RNA-cleaving DNA enzymes, were found to propel themselves by "burning" their RNA-coated track in the presence of metal cations.

In this paper, we describe the construction of a synthetic nanomotor that moves in a biased manner along a filament track using a variant of the burnt-bridge mechanism. The motor is an oligomer and consists of three linked coarse-grained units or beads, with catalytic and noncatalytic beads at two ends that are responsible for directed motion. Unlike the specific form of the link-burning event that gives rise to directed motion in the burnt-bridge model discussed above,^{27,28} the direction of motion of the oligomeric motor is determined by the asymmetric binding properties of the end beads of the motor. Unlike biomolecular motors, e.g., kinesins and myosins, that walk along filament tracks by changing their conformations, our motor uses motor-catalyzed chemical reactions that occur

a)Electronic mail: mjhuang@chem.utoronto.ca

^{b)}Electronic mail: rkapral@chem.utoronto.ca

on the filament in order to move. In contrast to many other studies of molecular motors, our mesoscopic description of motor dynamics includes an explicit particle-based specification of the motor geometry and the filament on which the motor moves, the solvent in which it resides, and, particularly, the chemical reactions that power its motion. Because such a detailed description of the entire system is adopted, the dynamical properties of the motor can be investigated in detail and, as well, it can be used as a testing platform for the design of synthetic molecular motors that could guide experiment.

In Sec. II, we describe, in qualitative terms, the geometry and properties of the motor and filament, the mechanism by which the motor moves on the filament, and a suggestion for how such motors could be realized in the laboratory. Section III provides a full microscopic description of the system including the specification of the intermolecular interactions and chemical reactions that supply the energy for directed motion. Section IV discusses how the dynamics of the system is followed and gives a quantitative description of the dynamical properties of the motor, including its velocity, mean square displacement (MSD), and operation under a load. Section V describes an analytically tractable stochastic model that is able to capture the main features of the simulation results. The conclusions of the paper are given in Sec. VI.

II. OLIGOMERIC MOTOR AND MECHANISM FOR ITS MOTION ON THE FILAMENT

The oligomeric motor comprises three beads linked in a linear arrangement. The end beads are denoted according to their function as catalytic M_C and reflector M_R beads; these beads are connected by a neutral M_N bead. Motors with more than one neutral bead can be constructed but we restrict our study to a three-bead oligomer. This motor binds to a filament that is also composed of beads linked to form a stiff polymer chain. The filament beads are of two different types called F_A and F_B . A sketch of the motor and filament is shown in Fig. 1. The motor and filament reside in a solution containing solvent fuel S_A and waste S_B species. When the motor M_C



FIG. 1. The motor consists of three beads: catalytic bead (M_C , red), neutral bead (M_N , gray), and reflector bead (M_R , blue). A fuel particle (S_A , orange) triggers the reaction on a substrate bead (F_A , purple) producing a product bead (F_B , black) and a waste bead (S_B , green). The arrow indicates the direction of propulsive motion.

bead is bound to a filament F_A bead, it is able to catalyze the reaction $F_A + S_A \rightarrow F_B + S_B$ that consumes a fuel molecule S_A in order to convert the filament F_A bead to an F_B bead and, in the process, yields a waste molecule S_B in the environment.

The mechanism giving rise to biased motion of the motor on the filament is easily understood if we consider the case where both the catalytic and reflector beads bind with equal strengths to the F_A filament beads, while the reflector bead is bound more weakly than the catalytic bead to the F_B filament beads. Suppose that the filament comprises only F_A beads initially. At some later time, the catalytic reaction that converts $F_A \rightarrow F_B$ with consumption of a fuel molecule will occur, leading to a configuration like that shown in Fig. 1. Due to Brownian motion, at a yet later time, the reflector bead will find itself in registry with an $F_B F_A$ pair of filament beads that was produced by the catalytic reaction. In this configuration, due to the variation of binding affinities across the $F_B F_A$ interface, the component of the net force on this motor bead along the filament direction is non-zero and directed to the right in Fig. 1. This interface behaves effectively as a barrier that *reflects* the M_R bead and also the entire oligometric motor because its beads are linked. After such a "reflection" event, the motor will undergo biased diffusion to a region containing fresh substrate F_A beads. Subsequent reactions will cause the process to repeat and the motor will continue to advance along the filament, as long as there are regions of fresh F_A to the right.

While the oligomeric motor described above was constructed to illustrate some of the generic features that could give rise to the directed motion of a molecular motor along a filament without invoking motor conformational changes, it is nevertheless useful to suggest how such motors might be made. For example, one may utilize a design strategy that is based on the DNA enzyme motors that move on a RNA substrate.³⁶ Specifically, the M_C bead can be synthesized with DNA single strands each of which consists of a catalytic core, whereas the noncatalytic M_R bead can be constructed by anchoring DNA single strands without catalytic activity. Fuel particles, S_A , can be treated as metal cations in the bulk solution using the fact that the DNA enzyme activity is dependent on the presence of metal cations.^{35–37} Binding of the motor beads to the filament beads may be realized by anchoring RNA single strands on an F_A substrate bead. Reactions catalyzed by DNA enzyme strands on the M_C bead will yield F_B product beads consisting of cleaved RNA strands, along with S_B waste particles. To ensure that motors located at the substrate-product interface have higher probability of moving toward the region with fresh substrates, the DNA strands on M_R bead should be engineered such that the binding affinity to the cleaved RNA strands is much weaker than that of the base pairs between DNA enzyme and cleaved RNA strands. The set of DNA-RNA base pairs could be designed such that the interaction energies satisfy the conditions discussed above for motor motion, or a similar set of conditions, that lead to biased forces. Note that while in such a realization of the motor the DNA and RNA strands attached to the motor beads undergo conformational changes, the three backbone beads in the oligomer do not change their conformation.

III. THE MOTOR, FILAMENT, AND REACTIVE ENVIRONMENT

Here, we present a specific realization of an oligomeric motor with the properties discussed above, along with a description of the solution in which it resides, and discuss in more detail how the dynamics of the motor and its environment are simulated.

Both the motor and filament tracks are linear chains of beads, where neighboring beads are linked by bonds described by harmonic springs, $V_{\text{bond}} = \frac{1}{2}k_s(r - r_0^{\alpha})^2$, where k_s is the spring constant, and r_0^{α} is the equilibrium length of the motor $\alpha = M$ or filament $\alpha = F$ bonds. The bending stiffness of the motor is included by adding an elastic spring connecting the first and last beads, with an equilibrium length of $2 r_0^M$. The bending stiffness of the filament is modeled by a three-body potential, $V_{\text{bend}} = k_b [1 - \cos(\phi - \phi_0)]$, where k_b is the bending energy, ϕ_0 is the equilibrium angle, and $\cos \phi = \hat{r}_{i-1,i} \cdot \hat{r}_{i,i+1}$, where $\hat{r}_{ij} = (\mathbf{r}_i - \mathbf{r}_j)/|\mathbf{r}_i - \mathbf{r}_j|$. In order to mimic experimental situations where filaments are usually immobilized by supporting matrices, the center of mass of the filament is fixed in space by applying an additional force to eliminate filament translation.³⁸

The solution surrounding the motor and filament comprises $N_S = N_{S_A} + N_{S_B}$ particles of S_A and S_B species. Inert solvent species could also be included that will dilute the concentrations of the fuel and waste molecules but we shall not consider that situation here. These chemical species interact with the motor and filament beads through central intermolecular potentials, $V_{\alpha\alpha'}(r)$, where $\alpha = M_C, M_N, M_R, F_A, F_B$ and $\alpha' = S_A, S_B$. The solvent species interact among themselves through multiparticle collisions³⁹⁻⁴² rather than intermolecular potentials. Multiparticle collision dynamics combines effective multiparticle collisions at discrete time intervals τ with streaming between two consecutive collisions. In order to carry out such collisions, particles are sorted into cubic cells ξ with linear size a. We employ the momentum-conserving Anderson thermostat version of multiparticle collision dynamics⁴³ where the postcollision velocity of particle *i* in cell ξ is given by

$$\mathbf{v}_{i}' = \mathbf{V}_{\xi} + \mathbf{v}_{i}^{\mathrm{ran}} - \sum_{j \in \mathrm{cell} \ \xi} \mathbf{v}_{j}^{\mathrm{ran}} / N_{\xi}, \tag{1}$$

and the components of $\mathbf{v}_i^{\text{ran}}$ are Gaussian random numbers with zero mean and variance $k_B T/m$, N_{ξ} is the total number of solvent particles in the cell, and \mathbf{V}_{ξ} is the center-of-mass velocity of these N_{ξ} particles. Grid shifting was employed to insure Galilean invariance.^{44,45}

The various species also participate in chemical reactions involving the motor and in the bulk of the solution. When the motor moves on the filament, at each time step the M_C bead is in contact with $N'_F = N'_{F_A} + N'_{F_B}$ filament beads comprising N'_{F_A} substrate F_A beads and N'_{F_B} product F_B beads. Each of these filament beads forms a complex with the M_C bead denoted by $(M_C F_A)$ or $(M_C F_B)$, and catalytic reactions,

$$(M_C F_A) + S_A \rightleftharpoons (M_C F_B) + S_B, \tag{2}$$

take place independently on each of the complexes. These reactions depend on the numbers of fuel N'_{S_A} and waste N'_{S_B} (N'_S = $N'_{S_A} + N'_{S_B}$) particles in the vicinity (within the interaction

TABLE I. The upper part of the table lists the average number of F_A (N'_{F_A}) , F_B (N'_{F_B}) , and total $(N'_F = N'_{F_A} + N'_{F_B})$ filament beads that form complexes with the M_C bead. The bottom part of the table compares the rate of production of F_B beads, \mathcal{R} , obtained from simulation \mathcal{R}^S and from theory \mathcal{R}^T (see Sec. V) for irreversible reactions on the filament and in the bulk.

$p_1(10^{-4})$	0.01	0.03	0.15	0.3	0.5	0.7	0.85	1
N'_{F_A}	1.99	2.00	1.69	1.52	1.43	1.31	1.19	1.17
N'_{F_B}	0.20	0.19	0.50	0.68	0.77	0.89	1.00	1.02
N'_F	2.19	2.19	2.19	2.20	2.20	2.20	2.20	2.19
$\mathcal{R}^{S}(10^{-4})$	0.072	0.18	0.86	1.55	2.41	3.11	3.59	3.64
$\mathcal{R}^{T}(10^{-4})$	0.073	0.22	0.93	1.67	2.63	3.38	3.74	4.31

range) of a single F_A or F_B bead in $(M_C F_A)$ or $(M_C F_B)$ complexes, respectively. Each of the N'_{S_A} particles has a probability per unit time p_1 of reacting to form products, so that the forward reaction rate is $\mathcal{R}_f = (dN_{F_B}/dt)_f = -(dN_{F_A}/dt)_f$ = $p_1N'_{F_A}N'_{S_A}$. Similarly, the reverse reactions occur with a probability per unit time of p_{-1} and the reverse reaction rate is given by $\mathcal{R}_r = p_{-1}N'_{F_B}N'_{S_B}$, so that the net reaction rate is $\mathcal{R} = \mathcal{R}_f - \mathcal{R}_r$. In the absence of the catalytic bead, these reactions are assumed to occur at a negligible rate and are not considered. The $(M_C F_A)$ and $(M_C F_B)$ complexes dissociate whenever the distance between the M_C bead and the filament bead in a complex is larger than their interaction range.

The concentrations of fuel and waste species are controlled by reactions, $S_B \stackrel{k_2}{\underset{k_2}{\Longrightarrow}} S_A$, that take place in a region \mathcal{B} of the bulk solution removed from the motor using the reactive version of multiparticle collision dynamics.^{46,47} Further information concerning the algorithm that was used to simulate the dynamics of the entire system, along with a specification of the system parameters, is given in Appendix A.

If the bulk-phase reaction rates are large and dominate those at the motor, a steady state involving these species is established quickly so that $N_{S_A}/N_{S_B} = k_2/k_{-2}$, and the ratio $Q = N_{S_A}/N_S = k_2/(k_2 + k_{-2})$ measures the fraction of fuel particles in the bulk. We may then write the forward reaction rate as $\mathcal{R}_f = p_1 N'_{F_A} N'_S (N'_{S_A}/N'_S) \approx p_1 N'_{F_A} N'_S (N_{S_A}/N_S)$ $= p_1 N'_{F_A} N'_S Q$. In a similar manner, the reverse rate can be written as $\mathcal{R}_r \approx p_{-1} N'_{F_D} N'_S (1-Q)$. The values of the average species numbers that participate in the motor reactions, along with the net reaction rate, are given in Table I for several values of p_1 using $p_{-1} = 10^{-6}$ and Q = 1. The average number of fuel plus waste particles that interact only with one of N'_{F_A} substrate beads is $N'_S \simeq 3.7$, which is approximately independent of p_1 . Since Q = 1, these are all fuel particles. Using these data, we can estimate the reaction rate \mathcal{R} from the formulas given above, and these estimates, also presented in Table I, are in good agreement with the direct simulation data.

IV. SIMULATION OF MOTOR MOTION

Figure 2 (Multimedia view) shows an example of the motion of the motor along the filament when the reverse motor reactions occur at a negligible rate. At time t = 0, the motor starts at the left end of the filament track. Reactions catalyzed



FIG. 2. Instantaneous motor and filament configurations at times t = 0, 30 000, 60 000, and 90 000. The motor beads are M_C (red), M_N (gray), and M_R (blue), and its position is indicated by an arrow. The filament beads are F_A (purple) and F_B (black). The reaction probabilities are $p_1 = 1.5 \times 10^{-5}$, $p_{-1} = 10^{-6}$, and Q = 1. (Multimedia view) [URL: http://dx.doi.org/10.1063/1.4922926.1]

by the M_C bead convert filament substrate F_A beads (purple) to product F_B beads (black). The motor randomly diffuses on the filament until it encounters an existing product bead. It is then reflected by the bead and moves to the right where there are fresh filament substrate F_A beads. In the course of its evolution, the motor catalyzes further reactions producing additional F_B filament beads and the reflection process continues. After time $t = 90\,000$, the motor is found to have moved a considerable distance to the right on the filament. The motor will continue its biased motion in this direction provided there are only F_A beads to the right: the nonequilibrium state of the filament and the reflection mechanism give rise to the directed motion.

The *x*-component of the M_R bead position on the filament, $R_x(t)$, is shown in Fig. 3. The motor randomly diffuses along the filament until the M_R bead encounters an F_BF_A interface. (The positions of F_B beads are indicated by red dots in the figure.) One can see that the motion of the motor is restricted when the M_R bead is in contact with an F_B bead. These contacts prevent R_x from decreasing and result in biased motor motion. Recall that the interaction strengths between the M_C bead and filament beads are chosen so that M_C bead can move across (or penetrate) F_B beads without experiencing reflecting forces. Therefore, catalytic reactions may occur when the M_C bead moves to the left of existing F_B beads. This process will produce "ineffective" barriers, indicated by the green open

TABLE II. Comparison of motor speed (*V*), average distance (Δx), and duration (Δt) between two consecutive effective barriers obtained from the simulation (*S*) and theory (*T*).

$p_1(10^{-4})$	0.01	0.03	0.15	0.3	0.5	0.7	0.85	1
$V^{S}(10^{-4})$	2.10	2.40	4.0	5.53	6.67	7.19	7.46	6.98
$V^{T}(10^{-4})$	1.51	2.44	4.28	5.29	6.23	6.83	7.09	7.48
$\begin{array}{l} \Delta x^S \\ \Delta x^T \end{array}$	13.4	11.24	5.64	4.91	3.69	3.27	3.05	2.76
	20.01	10.88	4.66	3.24	2.44	2.07	1.94	1.77
$\Delta t^{S}(10^{4})$ $\Delta t^{T}(10^{4})$	4.24	3.89	1.32	0.83	0.56	0.42	0.40	0.39
	13.63	4.53	1.07	0.60	0.38	0.30	0.27	0.23

squares in Fig. 3. These barriers have no effect on the motor dynamics.

The velocity of the motor was computed from the slope of the average value of $R_x(t)$ over 50 realizations of the dynamics and is given in Table II as a function of p_1 . As expected, the velocity increases as the forward catalytic reaction rate increases since this produces F_B filament beads that contribute to the reflection mechanism.

In Fig. 3, we see that as the motor executes biased diffusion on the filament track, it leaves as set of "footprints," the product filament beads. From information contained in 50 realizations of the dynamics, we can examine how the reaction rate influences the separation time (Δx) and duration time (Δt) between two reactions that produce effective barriers. These results are listed in Table II. As p_1 increases, both Δx and Δt first decrease quickly, followed by a slow decrease. For small p_1 , the waiting time for next reaction is long and the motor diffuses over large distances; however, when p_1 is large, reactions rapidly occur once M_C encounters an F_A bead that is near to an F_B bead produced in a previous reaction, and this shortens the separation Δx .

The short-time diffusive motion and the approach to the long-time ballistic motion of the motor on the filament are also evident in the plots of the MSD for various values of p_1 shown in Fig. 4. For instance, for $p_1 = 1.5 \times 10^{-5}$, at short times $MSD(t) \approx 0.01t^{0.93}$ so that the diffusion coefficient is $D = 5 \times 10^{-3}$, which is comparable to its value when there are no catalytic reactions ($D = 4 \times 10^{-3}$). For long times, a fit to the data yields $MSD(t) \approx 5.38 \times 10^{-6}t^{1.7}$ with the exponent



FIG. 3. The motor trajectory, R_x versus t, on the filament. The symbols denote filament product beads: the red dots are effective barriers, and the green squares are ineffective barriers. The displacement (Δx) and duration (Δt) between two effective barriers are indicated.



FIG. 4. Log-log plots of the motor MSD versus time for various values of p_1 . The dashed and dotted lines are the fits for the ballistic and diffusive regimes, respectively.

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TABLE III. Comparison of motor speed from simulation (V^S) and theory (V^T) as a function of Q and of p_{-1} . Upper part of the table compares motor velocity from simulations with $p_1 = 10^{-4}$ and $p_{-1} = 10^{-6}$ and theory with an irreversible filament reaction ($p_1 = 10^{-4}$ and $p_{-1} = 0$) as a function of Q. Lower part of the table compares motor velocity from simulations and theory with reversible filament reaction using Q = 0.5 and (a) $p_1 = 10^{-4}$, (b) $p_1 = 10^{-5}$, and (c) $p_1 = 10^{-6}$ as a function of p_{-1} .

-								
	Q	0.02	0.1	0.3	0.5	0.7	1	
	$V^{S}(10^{-4})$ $V^{T}(10^{-4})$	1.37 2.10	3.90 3.80	5.75 5.40	6.28 6.24	6.71 6.79	6.98 7.31	
	$p_{-1}(10^{-4})$	0.01	0.05	0.1	0.3	0.5	0.8	0.1
(a)	$V^{S}(10^{-4})$ $V^{T}(10^{-4})$	6.22 6.08	5.66 5.92	6.44 5.97	6.23 5.91	5.58 5.80	5.72 5.86	5.77 5.86
(b)	$V^{S}(10^{-4})$ $V^{T}(10^{-4})$	2.91 2.90	2.83 2.86	3.16 2.86	2.89 2.85	2.74 2.83	2.18 2.85	2.49 2.83
(c)	$V^{S}(10^{-4})$ $V^{T}(10^{-4})$	1.84 1.10	1.89 1.10	1.69 1.09	1.76 1.09	1.50 1.09	1.04 1.10	0.95 1.09

approaching a value of 2 corresponding to that for ballistic motion.

For completeness, Table III shows how the average motor velocity depends on Q, which controls the fuel concentration, and p_{-1} which determines the reverse motor reaction rate. As expected, when Q increases so does the speed with the largest value obtained for Q = 1 corresponding to all fuels. The motor velocity does not depend strongly on the reverse reaction rate and decreases only weakly with its increase. The small magnitude of the decrease is due to the fact that the barrier-removing process is diffusion-controlled: in order to remove a barrier from the filament, the motor needs to diffuse to a position where the M_C bead is in contact with an F_B bead and a $(M_C F_B)$ complex forms. Once such a configuration is attained,

the motor must wait for a reverse reaction to occur, but during this waiting time, the motor may diffuse away from the F_B leading to dissociation of the complex. Consequently, reverse motor reactions have a limited effect on motor dynamics.

A. Motor subject to an external force

Cargo transport is an important function of molecular motors, and to perform this task, a motor must be able to operate under an external load. We now investigate the effect of a load on the dynamics of our oligomeric motor by applying a force $-F\hat{x}$ on the M_N motor bead under the condition of $p_1 = 10^{-4}$ and Q = 1. The mean motor velocity was determined from averages over 50 realizations of the motor dynamics where, in each realization, the motor was subject to the force F. Figure 5(a) shows the motor velocity V (black circles) as a function of F, and we see that V decreases rapidly with increasing F. For values of F > 0.2, the velocity takes negative values so that $F_s \approx 0.2$ is the stall force. Insight into the nature of the motor motion for negative values of V can be gained from an examination of Fig. 5(b), which shows a sample trajectory taken from a simulation with F = 0.1. One can see that the motor is first localized at positions near x = 0 against a reflecting barrier. At time $t \approx 70\,000$, the motor, which is pulled by the external force, moves backward until a new reflecting barrier produced by a catalytic reaction is reached.

Each reflecting barrier has a finite energy depth, and the probability of hopping across the barrier increases with the external force strength. By examining the simulation trajectories in detail, we found that the probability of observing barrier crossing events increases with external load: when F = 0.1, 2% of the trajectories experience barrier crossing events, while for F = 0.5, the percentage increases to 96%. Due to the prevalence of barrier crossing events for large F, backward



FIG. 5. (a) Motor velocities with (interpolated black circles) and without (blue diamonds) barrier crossing events. The theoretical prediction of the motor velocity for various values of the external force is shown as red squares. The dotted line indicates V = 0. (b) A sample trajectory under an external force F = 0.1 showing a barrier crossing event at approximately t = 70000. (c) Average number of filament beads that encounter the M_C bead versus the external force: N'_{F_A} (black circles) and N'_{F_B} (red squares). (d) Motor velocity as a function of the external force without chemical reactions. The slope of the linear fit (red line) yields the inverse friction coefficient $\zeta^{-1} \simeq 0.021$.

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motor motions induced by the external force dominate over forward propulsive motions powered by catalytic reactions. To determine the motor velocity in the absence of barrier crossing events, only the trajectories with positive velocities were used to determine the average motor velocity. These results are shown as blue diamonds in Fig. 5(a). One can see that the motor velocity decreases and eventually tends to zero when Fapproaches the stall force that prevents the motor from moving across the reflecting barrier represented by an F_B bead.

V. STOCHASTIC MODEL FOR MOTOR SPEED

A simple stochastic model is able to capture many of the essential features of the motor dynamics observed in the simulations. While a similar model with only irreversible reactions was previously studied,²⁷ in this section we consider a general stochastic model with irreversible reactions on the filament in the presence of an external load. In Appendix B, the generalization of the model to include reverse reactions is described.

First, we consider an irreversible motor reaction with $p_{-1} = 0$, and a reversible reaction in the bulk characterized by various values of Q. As shown in Fig. 6, the motor diffuses along the *x*-axis starting at t = 0 with the M_C bead at x = 0 and the M_R bead at $x = L_M$, where $L_M = 4$ is the motor length given by the distance between the centers of the M_C and M_R beads. At time t = 0, a catalytic reaction occurs producing a product bead at the position of M_C bead. The distance between the surface of M_R bead at $M_N M_R$ interface and the F_B bead is $L \approx 3$. We assume that the M_R bead begins to interact with the F_B bead when the motor moves a distance L to the left. Given the initial position of the $M_N M_R$ interface x'(t = 0) = L, the probability distribution function $\phi(x',t)$ of x' when the motor is subject to an external force $-F\hat{x}'$ satisfies the diffusion equation,

$$\frac{\partial \phi}{\partial t} = D \frac{\partial^2 \phi}{\partial x'^2} + \frac{F}{\zeta} \frac{\partial \phi}{\partial x'},\tag{3}$$

where *D* is the motor diffusion constant, and ζ is the friction coefficient. By solving Eq. (3) with a reflecting barrier condition, $\partial_{x'}\phi = 0$ at x' = 0, the probability of finding the



FIG. 6. Schematic representation of the motor under an external force (dashed arrow, $-F\hat{x}$) which acts on the M_N bead (gray). The solid line is the center of the F_B bead at x = 0 and indicates the position of a reflecting barrier for motor propulsion, and dotted lines are the center and the interaction boundary of the M_R bead at $x = L_M$ and x = L, respectively. The solid arrow indicates the direction in which the motor moves.

 $M_N M_R$ interface at a position between x' and x' + dx' at time t is

$$\phi(x',t)dx' = \frac{1}{\sqrt{4\pi Dt}} \left[e^{-\frac{(x'+ut+L)^2}{4Dt}} + e^{-\frac{(x'+ut-L)^2}{4Dt}} \right] dx', \tag{4}$$

where $u = F/\zeta$. The probability of a forward reaction taking place during time *t* and *t* + *dt* is given by

$$f(t)dt = Ap_1 e^{-Ap_1 t} dt, (5)$$

where $A = N'_{F_A}N'_SQ$ accounts for the effects from the filament configuration and number of fuel particles involved in the reaction.

Given a reflecting barrier created at time t = 0 at position x' = 0, the average position of a new effective barrier created at the position of the M_C bead, x' - L, is given by

$$\Delta x = \int_{t=0}^{\infty} \int_{x'=L}^{\infty} (x'-L)\phi(x',t)dx'f(t)dt,$$
 (6)

where the spatial integration is taken in the interval $x' = [L, \infty]$ since only the product bead produced in the region x > 0 is effective for motor propulsion, i.e., the position of the $M_N M_R$ interface must lie in the region where x' > L so that x' - L > 0. Substituting Eqs. (4) and (5) into Eq. (6), one gets

$$\Delta x = \left[\left(\frac{\sqrt{D}}{2} + \frac{u^2}{4Ap_1\sqrt{D}} \right) \left(Ap_1 + \frac{u^2}{4D} \right)^{-\frac{1}{2}} - \frac{u}{2Ap_1} \right] \\ \times \left[1 + \exp\left(\frac{-Lu}{D} - 2L\sqrt{\frac{u^2}{4D^2} + \frac{Ap_1}{D}} \right) \right].$$
(7)

The average time interval between two reactions is

$$\Delta t = \int_{t=0}^{\infty} f(t)dt = \frac{1}{Ap_1},\tag{8}$$

and thus the average speed is given by $V = \frac{\Delta x}{\Delta t}$.

A. No external force

In the absence of an external force, $u = F/\zeta = 0$, and Eq. (7) reduces to

$$\Delta x = \frac{1}{2} \sqrt{\frac{D}{Ap_1}} \Big[1 + e^{-2L\sqrt{Ap_1/D}} \Big],$$
(9)

yielding a positive motor velocity,

$$V = \frac{1}{2}\sqrt{Ap_1D} \bigg[1 + e^{-2L\sqrt{Ap_1/D}} \bigg].$$
 (10)

Using the simulation values of the parameters, $N'_S Q \simeq 3.7$, $D \simeq 4 \times 10^{-3}$, $L \simeq 3$, and N'_{F_A} for Q = 1 given in Table I, one can compute Δx and Δt using Eqs. (9) and (8), respectively, and, thus, the motor velocity, V. These theoretical values of Δx , Δt , and V are given in Table II. In addition, one can also compute V for different fuel concentrations controlled by Q, which enters in the expression for A. Table III compares these theoretical results with those from direct simulation of the motor dynamics. Good agreement is found.

B. With an external force

In order to obtain an analytical estimate for the motor velocity in the presence of an external force, two parameters

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are necessary: the number N'_{FA} as a function of F and friction coefficient ζ . The numbers N'_{FA} as a function of F can be determined from an analysis of the motor-filament configurations obtained in the simulations in Sec. IV A, and these results are shown in Fig. 5(c). The friction coefficient, ζ , was computed from a linear fit of the plot of velocity versus force shown in Fig. 5(d) in simulations without chemical reactions. This fit yields $\zeta^{-1} \simeq 0.021$. The estimate of the motor velocity may then be computed using Δx from Eq. (7) and Δt from Eq. (8). These theoretical results are compared with simulations in Fig. 5(a), and we find that the theory is able to capture the trends in the data.

VI. CONCLUSION

This study provided an example of a synthetic nanomotor which is able to execute directed motion on a filament using a variant of the burnt-bridge mechanism. The motor exploits the different binding energies of the motor groups to the filament segments and catalytic reactions with the filament to which it bound. As discussed in the text, one way in which such motors could be realized in the laboratory is by designing the motor and filament to be made of beads with anchored DNA enzyme strands and substrate RNA strands, respectively, and by utilizing the activity of DNA strands on different motor beads to implement a propulsion mechanism like that described in this paper. Such a design strategy spontaneously breaks the leftright symmetry of the direction of motion without predefining the "burn" event to bias the motion in a specific direction. Of course, there are many ways to introduce motor reactions to drive the motion; some of which need not involve the bulkphase reactions. However, the use of fuel particles in the bulk solution allows us to control motor velocity remotely, similar to what has been suggested in the experiments.³⁶ The motor geometry can also be modified as needed to accomplish specific tasks.

Many biological molecular motors evolved to deliver cargo to specific targets, and they usually accomplish this by moving along filament tracks so that orientational Brownian motion is suppressed. Our synthetic motor uses this strategy and we have demonstrated that it is able to function under an external load while moving on the filament. Previous studies of motor motion used burnt-bridge models that involved only stochastic jumps and burn events.^{27,28} Through microscopic simulations and stochastic modeling, we showed how fuel concentration and reaction conditions determine the velocity and properties of the motor motion on the filament. The use of multiparticle collision dynamics preserves hydrodynamics, which can be used for further studies of hydrodynamic mediated collective motion of burnt-bridge motors. Thus, this work provides a modeling platform for the investigation of synthetic motor design and dynamics on filament tracks under various conditions.

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APPENDIX A: SIMULATION PARAMETERS

Simulations of the dynamics of the oligometric motor are carried out in a $100 \sigma \times 20 \sigma \times 20 \sigma$ box with periodic boundaries. All motor beads have the same size with diameter d_{M_C} $= d_{M_N} = d_{M_R} = 2\sigma$, while the diameters of the filament beads are $d_{F_A} = d_{F_B} = 1.5 \sigma$. For small motors, exclusion forces are important and these are sufficient to bind the motor to the filament. Since these attractive exclusion mean-force potentials are strongest when a motor bead resides at the junction between two filament beads (cf. Fig. 1), this leads to a periodic mean-force potential along the filament. In order to tune the magnitude of this effective potential and control the binding properties of the motor beads to the different types of filament beads, we supplement these effective potentials with repulsive Lennard-Jones potentials for the interactions between the motor and filament beads. The repulsive Lennard-Jones potentials have the form, $V(r_{ij}) = 4 \epsilon_{\alpha\alpha'} [(\sigma_{\alpha\alpha'}/r_{ij})^{12}]$ $-(\sigma_{\alpha\alpha'}/r_{ij})^6 + 1/4]\theta(r_c - r)$, where $\theta(r)$ is the Heaviside function, and the separation of particle *i* of type α from particle *j* of type α' is $r_{ij} = |\mathbf{r}_i - \mathbf{r}_j|$, where $\alpha, \alpha' \in \{F_A, F_B, M_C, M_N, d_N\}$ M_R . The interaction between two beads has strength $\epsilon_{\alpha\alpha'}$ and vanishes when the separation is larger than the cutoff distance $r_c = 2^{1/6} \sigma_{\alpha \alpha'}$, taking $\sigma_{\alpha \alpha'} = (d_{\alpha} + d_{\alpha'})/2$. The point particle solvent species, S_A and S_B , also interact with the motor and filament beads through repulsive Lennard-Jones potentials. The interaction strength is $\epsilon_{S\alpha}$, and the cutoff distance is $r_c = 2^{1/6} \sigma_{S\alpha}$, where $\sigma_{S\alpha} = (d_S + d_\alpha)/2$ and $d_S = \sigma$ is the effective diameter of the S species when it interacts with a motor or filament bead. All solvent species have the same mass m, whereas masses of the motor and the filament beads are chosen to be $m_{\alpha} = (d_{\alpha}/d_S)^3 m$ so that they have the same mass density as a solvent particle. The streaming step of the dynamics is governed by molecular dynamics under these potential functions. The solvent collisions are carried out at times τ using reactive multiparticle collision dynamics as discussed above. The bulk-phase region $\mathcal B$ where reactions take place is defined by $r^2 = r_y^2 + r_z^2 > 10^2$.

Other pertinent parameters used in the simulations are the molecular dynamics time step, $\delta t = 0.01 t_0$, the collision time, $\tau = 0.5 t_0$, and the temperature of the system, $k_B T = 0.2 \epsilon$. A solvent particle interacts with a motor or filament bead with interaction strength $\epsilon_{S\alpha} = \epsilon$, whereas the ϵ values for interactions among motor and filament beads are $\epsilon_{F_BM_R} = \epsilon$, and otherwise $\epsilon_{\alpha\alpha'} = 0.1 \epsilon$. Note that there are no repulsive interactions among species of the same type; i.e., ϵ_{MM} $= \epsilon_{FF} = \epsilon_{SS} = 0$. There are $N = 358\,422$ solvent particles, which give a solvent density of $n_0 \approx 9$. The multiparticle collision cell size is $a = \sigma$. Two neighboring beads in the motor or in the filament are linked by springs with spring constant k_s $= 100 \epsilon / \sigma^2$ and with equilibrium lengths $r_0^M = 2 \sigma$ and $r_0^F = \sigma$. The bending stiffness of the filament is $k_b = 100 \epsilon$ and the equilibrium angle is $\phi_0 = 0$. Results are reported in dimensionless units based on mass *m*, energy ϵ , time $t_0 = \sqrt{m\sigma^2/\epsilon}$, and length σ .

APPENDIX B: REVERSIBLE CATALYTIC MOTOR REACTION

When the motor moves on the filament, it creates effective and ineffective barriers. While ineffective product beads may be ignored for irreversible catalytic reactions, they become important when conversion of effective product beads back into substrate beads occurs. Here, we construct a simple model, illustrated in Fig. 7, to estimate the motor speed taking the effects of reverse reactions into account. The diagram shown in Fig. 7 comprises nine filament configurations connected by five paths. The sequence of configurations in each path starts with the configurations in the left column and ends with the configurations containing a new effective barrier shown in the right column. To calculate the probability of each path, we consider the average probabilities, p_{-} for $(a) \rightarrow (c)$ and (b) \rightarrow (d) and p'_{-} for (d) \rightarrow (e), of removing an effective barrier, while the probabilities of producing a new effective barrier are $p_+ = 1 - p_-$ for $(a) \rightarrow (g)$ and $(b) \rightarrow (g)$ and $p'_+ = 1 - p'_-$ for $(d) \rightarrow (h)$. Thus, the probabilities of each path can be written as

$$(a) \to (c) \to (f) : p^{(0)} = (1 - p_0)p_-,$$

$$(a) \to (g) + (b) \to (g) : p^{(1)} = (1 - p_0)p_+ + p_0p_+,$$

$$(b) \to (d) \to (h) : p^{(2)} = p_0p_-p'_+,$$

$$(b) \to (d) \to (e) \to (i) : p^{(3)} = p_0p_-p'_-,$$

where p_0 is the probability of having an ineffective barrier at t = 0 indicated by Fig. 7(b). For each path, the average duration of a new effective barrier is estimated from the sum of the average times for each reaction, i.e.,

$$(a) \to (c) \to (f) : \vec{t}_{+}^{(0)} = \frac{1}{Ap_1} + \frac{1}{Bp_{-1}},$$
$$(a) \to (g) + (b) \to (g) : \vec{t}_{+}^{(1)} = \frac{1}{Ap_1},$$

$$(b) \to (d) \to (h) : \overline{t}_{+}^{(2)} = \frac{1}{Ap_{1}} + \frac{1}{Bp_{-1}},$$

$$(b) \to (d) \to (e) \to (i) : \overline{t}_{+}^{(3)} = \frac{1}{Ap_{1}} + \frac{2}{Bp_{-1}}.$$

In each final configuration, new effective barriers are denoted by red rods, and their positions are $\overline{x}_{+}^{(n)}$. Therefore, the motor speed can be estimated by

$$\overline{V} = \sum_{n=0}^{3} p^{(n)} \frac{\overline{x}_{+}^{(n)}}{\overline{t}_{+}^{(n)}}.$$
(B1)

Given the estimate of average duration of each path, to calculate average speed, one needs to compute the probabilities $(p^{(n)})$ and positions $(\overline{x}^{(n)}_+)$ in Eq. (B1). First, given an effective barrier at x = 0, we calculate the probability (p_0) of producing an ineffective barrier and its average position $x = -\overline{x}$. Note that an ineffective barrier will be created only when the $M_N M_R$ interface lies in the region 0 < x' < L, and, thus, the probability p_0 is

$$p_{0} = \int_{t=0}^{\infty} \int_{x'=0}^{L} \phi(x',t) f(t) dx' dt$$
$$= \frac{1}{2} \left[1 - e^{-2L\sqrt{Ap_{1}/D}} \right],$$
(B2)

and the average position of the ineffective barrier is

$$-\overline{x}' = \int_{t=0}^{\infty} \int_{x'=0}^{L} (x'-L)\phi(x',t)f(t)dx'dt$$
$$= \sqrt{\frac{D}{Ap_1}} \left[e^{-L\sqrt{Ap_1/D}} - \frac{1}{2} - \frac{1}{2}e^{-2L\sqrt{Ap_1/D}} \right].$$
(B3)

Second, we calculate the probability of removing an effective barrier in between the M_R bead and an ineffective barrier. To remove the barrier, the M_R bead must be in a certain region on the filament so that the M_C bead is in contact with an existing effective barrier. Consider a case where the initial and final positions of the $M_N M_R$ interface are x_i and x_f , respectively, and an effective barrier is at $x = x_f - L$. The probability of finding the interface located between x_f and $x_f + \delta x$ at



FIG. 7. Paths creating new barriers. The filament configuration is represented by solid bars, where existing barriers (black bars), ineffective barriers (green dotted-dashed bars), new barrier (red bars), and removed barriers (gray bars) are plotted. The arrows indicate the transition direction to the next filament configuration as well as the associating transition probabilities.

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time t is

$$\phi'(x_i, x_f, t)\delta x = \frac{\delta x}{\sqrt{4\pi Dt}} \left[e^{-\frac{(x_f - x_i)^2}{4Dt}} + e^{-\frac{(x_f + x_i)^2}{4Dt}} \right],$$
(B4)

and, thus, the probability of a reverse reaction taking place when the M_C bead encounters an effective barrier is

$$\tilde{p}(x_i, x_f) = \int_{t=0} \delta x \phi'(x_i, x_f, t) B p_{-1} e^{-B p_{-1}} t dt$$

$$= \frac{\delta x}{2} \sqrt{\frac{B p_{-1}}{D}} \left[e^{-|x_f - x_i| \sqrt{\frac{B p_{-1}}{D}}} + e^{-|x_f + x_i| \sqrt{\frac{B p_{-1}}{D}}} \right],$$
(B5)

where $Bp_{-1}dt$ is probability of a reverse reaction occurring between time t and t + dt and $B = N'_{F_B}N'_S(1 - Q)$. Using Eq. (B5), the probability of removing the barrier at x = 0 in configuration (b) is

$$p_{-} = \tilde{p}(x_i = L, x_f = L).$$
 (B6)

To calculate the removal probability p'_{-} in configuration (d), one needs to shift the coordinate so that the barrier (the green dotted-dashed bar) is at x = 0, and the initial position of the M_R bead is then at $x = L + \overline{x}'$ with final position at x = L. Thus, we have

$$p'_{-} = \tilde{p}(x_i = L + \overline{x}', x_f = L).$$
(B7)

Third, we calculate the positions of new barriers shown in configurations (f)–(i). After barrier removal, in the paths (c) \rightarrow (f) and (e) \rightarrow (i), the motor dynamics evolves with an effective barrier located at $x = -\overline{x}$, whereas in the path (d) \rightarrow (h), the effective barrier is at $x = -\overline{x}'$. Here, we assume that there exists an effective barrier which was produced previously at the position $x = -\overline{x}$ (given by Eq. (9) in the irreversible case). To calculate the positions $\overline{x}_{+}^{(n)}$, we consider a case where the reflecting barrier is located at $x = -x_b$ and the initial position of the $M_N M_R$ interface is x_i . By shifting the reflecting barrier to the position x'' = 0, we have

$$\phi^*(x'',t) = \frac{1}{\sqrt{4\pi Dt}} \left[e^{-\frac{(x''-x_0)^2}{4Dt}} + e^{-\frac{(x''+x_0)^2}{4Dt}} \right], \quad (B8)$$

which satisfies the diffusion equation with the initial position of $M_N M_R$ interface at $x'' = x_0 = x_i + x_b$ and a reflecting barrier condition at x'' = 0. The average position of the new barrier can be computed by

$$\tilde{x}(x_b, x_i) = -x_b + \int_{t=0}^{\infty} \int_{x''=L}^{\infty} (x'' - L)\phi^*(x'', t)f(t)dtdx''$$

= $\overline{x}e^{-(x_i + x_b - L)\sqrt{Ap_1/D}} - L + x_i.$ (B9)

Using Eq. (B9), the new barrier positions can be computed

$$\overline{x}_{+}^{(0)} = \tilde{x}(x_{b} = \overline{x}, x_{i} = L),
\overline{x}_{+}^{(1)} = \tilde{x}(x_{b} = 0, x_{i} = L),
\overline{x}_{+}^{(2)} = \tilde{x}(x_{b} = \overline{x}', x_{i} = L),
\overline{x}_{+}^{(3)} = \tilde{x}(x_{b} = \overline{x}, x_{i} = L - \overline{x}').$$
(B10)

Taking the values of N'_{FA} and N'_{FB} from the simulations (shown in Table IV) and $\delta x = 1$ is chosen to be equal to the bond length between two filament beads, the probabilities $p^{(n)}$ can be computed by using Eqs. (B2), (B6), and (B7) as well

TABLE IV. The average numbers N'_{FA} , N'_{FB} , and their total N'_{F} of filament beads that form complexes with the M_{C} bead for Q = 0.5, various values of p_{-1} , and (a) $p_1 = 10^{-4}$, (b) $p_1 = 10^{-5}$, and (c) $p_1 = 10^{-6}$.

	$p_{-1}(10^{-4})$	0.01	0.05	0.1	0.3	0.5	0.8	1
(a)	N'_{F_A}	1.41	1.38	1.45	1.51	1.49	1.56	1.58
	N'_{F_P}	0.78	0.82	0.74	0.68	0.71	0.63	0.61
	N'_F	2.19	2.20	2.20	2.20	2.20	2.19	2.19
(b)	N'_{F_A}	1.90	1.88	1.92	1.95	1.96	2.00	2.00
	N'_{F_B}	0.30	0.31	0.28	0.24	0.23	0.19	0.20
	N'_F	2.19	2.19	2.19	2.19	2.19	2.19	2.19
(c)	N'_{FA}	1.99	2.04	2.00	2.04	2.06	2.08	2.08
	N'_{F_P}	0.20	0.15	0.19	0.15	0.13	0.11	0.11
	N'_F	2.19	2.19	2.19	2.19	2.19	2.19	2.19

as new barrier positions $\overline{x}_{+}^{(n)}$ and durations $t_{+}^{(n)}$. Substituting these quantities into Eq. (B1), one can estimate motor speed in the presence of both forward and reverse reactions. Table III presents these results.

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