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Stokes efficiency of molecular motors with inertia

Hongyun Wang

Department of Applied Mathematics and Statistics, University of California, Santa Cruz, CA 95064, United States

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ABSTRACT

A molecular motor utilizes chemical free energy to generate a unidirectional motion in a viscous media. The stochastic motion of a motor is governed by a Langevin equation coupled to the chemical occupancy state. The change of chemical occupancy state is governed by a discrete Markov process. The Stokes efficiency was introduced to measure how "efficiently" the motor uses chemical free energy to drive through the surrounding fluid. For the overdamping case where the effect of inertia is ignored, it was proved that the Stokes efficiency is bounded by 100% [H. Wang, G. Oster, The Stokes efficiency for molecular motors and its applications, Europhysics Letters 57 (2002) 134–140]. Here we present a proof for the general case.

1. Introduction and mathematical formulation

A molecular motor, in general, has many internal and external degrees of freedom. One of these degrees of freedom is associated with the motor's unidirectional motion, the main biological function of the motor. For example, a kinesin dimer walks along a microtubule filament toward the positive end [1,2]. There are many levels of models for molecular motors, from simple kinetic models with a few states to all atom molecular dynamics. In a modeling approach of intermediate level, the unidirectional motion is followed explicitly and the effects of other degrees of freedom are modeled in the mean field potential affecting the unidirectional motion [3–6]. To introduce this modeling approach of intermediate level, we consider the one-dimensional motion of a small object in a fluid medium and subject to a potential. The stochastic motion of the object is governed by Newton's second law:

$$\begin{aligned} \frac{\mathrm{d}\mathbf{q}}{\mathrm{d}t} &= -\zeta\frac{\mathbf{q}}{m} - \phi'(\mathbf{x}) + \sqrt{2k_BT\zeta}\frac{\mathrm{d}\mathbf{W}(t)}{\mathrm{d}t} \\ \frac{\mathrm{d}\mathbf{x}}{\mathrm{d}t} &= \frac{\mathbf{q}}{m} \end{aligned} \tag{1}$$

where $\mathbf{x}$ is the position, m is the mass, $\mathbf{q}$ is the momentum, and ζ is the drag coefficient of the object. $\phi(x)$ is the potential acting on the object and $\mathbf{W}(t)$ is the Wiener process. Here we adopt the convention that stochastic processes are denoted by boldface letters. The object is driven by three forces: (1) the drag force $-\zeta\mathbf{q}/m$, which is always opposing the motion, (2) the force derived from the potential $-\phi'(\mathbf{x})$, and (3) the Brownian force $\sqrt{2k_BT\zeta}\mathrm{d}\mathbf{W}(t)/\mathrm{d}t$. Both the drag force and the Brownian force are caused by the bombardments of surrounding fluid molecules. The amplitude of Brownian force is related to the drag coefficient as $\sqrt{2k_BT\zeta}$, which is a result of the fluctuation-dissipation theorem [7,8]. Here k_B is the Boltzmann constant and T the absolute temperature [9].

In a molecular motor, the potential is not static. Instead, the potential changes with the current chemical occupancy state of the motor. Let N be the number of possible chemical occupancy states of the motor in consideration. Let $\{1, 2, \ldots, N\}$ denote the set of N occupancy states, and $\{\phi_1(x), \phi_2(x), \ldots, \phi_N(x)\}$ denote the corresponding set of N potentials where $\phi_j(x)$

E-mail address: hongwang@ams.ucsc.edu.

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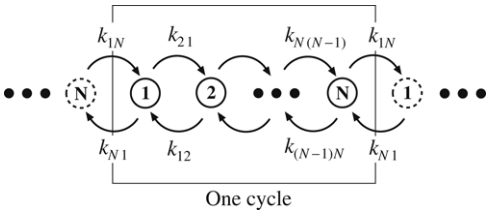

Fig. 1. Chemical reaction diagram of a hypothetical motor. Each reaction cycle consists of N chemical states. The transition rates and the corresponding transitions are shown in the diagram. In particular, k_{1N} is the rate of transition from state N of the current cycle to state 1 of the next cycle; and k_{N1} is the rate of transition from state 1 of the current cycle to state N of the previous cycle.

is the motor potential when the motor is in chemical occupancy state j. The mechanical motion of a motor is governed by the Langevin equation:

$$\begin{aligned} \frac{\mathrm{d}\mathbf{q}}{\mathrm{d}t} &= -f_0 - \zeta\frac{\mathbf{q}}{m} - \phi'_{\mathbf{S}}(\mathbf{x}) + \sqrt{2k_BT\zeta}\frac{\mathrm{d}\mathbf{W}(t)}{\mathrm{d}t} \\ \frac{\mathrm{d}\mathbf{x}}{\mathrm{d}t} &= \frac{\mathbf{q}}{m} \end{aligned} \tag{2}$$

where f_0 is a conservative load force on the motor, $\mathbf{S}(t)$ denotes the current chemical occupancy state and $\phi_{\mathbf{S}}(x)$ is the periodic motor potential corresponding to $\mathbf{S}(t)$. Notice that Langevin equation (2) is coupled to the current chemical occupancy state $\mathbf{S}(t)$. The periodic potential $\phi_{\mathbf{S}}(x)$ in (2) varies with $\mathbf{S}(t)$. When the motor switches to another occupancy state, the periodic potential $\phi_{\mathbf{S}}(x)$ in (2) changes accordingly. The change of chemical occupancy state $\mathbf{S}(t)$ is governed by a continuous time discrete space Markov process (a jump process):

$$Pr[\mathbf{S}(t+\Delta t) = j \mid \mathbf{S}(t) = i] = \Delta t k_{ji}(\mathbf{x}(t)) + o(\Delta t), \quad j \neq i \tag{3}$$

where $k_{ji}(x)$ is the rate of transition from occupancy state i to occupancy state j when the motor is at position x. Notice that the transition rate $k_{ji}(x)$ depends on the motor position x. As a result, in Markov process (3) the evolution of chemical state $\mathbf{S}(t)$ is affected by the motor position $\mathbf{x}(t)$. Conversely, in Langevin equation (2) the evolution of motor position $\mathbf{x}(t)$ is affected by the chemical state $\mathbf{S}(t)$. Thus, Langevin equation (2) and Markov process (3) are coupled, and together they govern the stochastic evolution of both the mechanical motion and chemical reaction of the motor. Let us focus on the case where in each cycle of the chemical reaction the motor goes sequentially through a set of N occupancy states. As illustrated in Fig. 1, for $1 < j < N$, from state j the motor can jump either to state $j+1$ or to state $j-1$ of the current cycle. From state 1, the motor can jump either to state 2 of the current cycle or to state N of the previous cycle. Similarly, from state N, the motor can jump either to state 1 of the next cycle or to state $N-1$ of the current cycle.

In experiments, only average quantities can be measured repeatedly and reliably. All average quantities can be calculated by following the probability density of the motor. Let us consider an ensemble of motors, each evolving in time independently and stochastically according to Langevin equation (2) coupled with the Markov process (3). Let $\rho_j(x, q, t)$ be the probability density that the motor is at position x, with momentum q and in occupancy state j at time t. Mathematically, $\rho_j(x, q, t)$ is introduced as

$$\rho_j(x,q,t) = \lim_{\substack{\Delta x\to 0\\ \Delta q\to 0}} \frac{Pr\begin{bmatrix} x \le \mathbf{x}(t) < x+\Delta x \\ q \le \mathbf{q}(t) < q+\Delta q, \end{bmatrix} \quad \mathbf{S}(t) = j}{\Delta x \Delta q} \tag{4}$$

and $\{\rho_j(x, q, t)\}$ is governed by the coupled Fokker–Planck equation corresponding to Langevin equation (2) and Markov process (3) [10]:

$$\frac{\partial \rho_j}{\partial t} = \frac{\partial}{\partial q}\left[\left(\phi'_j + f_0 + \frac{\zeta}{m}q\right)\rho_j\right] + k_BT\zeta\frac{\partial^2\rho_j}{\partial q^2} - \frac{\partial}{\partial x}\left[\frac{q}{m}\rho_j\right] + I_{j-1/2} - I_{j+1/2}, \quad j = 1, 2, \ldots, N \tag{5}$$

where $I_{j+1/2}$ is the net probability flux in the reaction direction from state j to state $j+1$. As shown in Fig. 1, state 0 refers to state N of the previous cycle and state $N+1$ refers to state 1 of the next cycle. $I_{j+1/2}$ is given by

$$\begin{aligned} I_{1/2} &= I_{N+1/2} \equiv k_{1N}\rho_N - k_{N1}\rho_1 \\ I_{j+1/2} &\equiv k_{(j+1)j}\rho_j - k_{j(j+1)}\rho_{j+1}, \quad 0 < j < N \end{aligned} \tag{6}$$

In Fokker–Planck equation (5), the transition rates $k_{ij}(x)$ cannot be arbitrarily specified. Instead these transition rates are constrained by the condition of detailed balance, which ensures that if the system is brought to an equilibrium, then the equilibrium solution is given by the Boltzmann distribution and the probability flux vanishes everywhere [9]. To derive the condition of detailed balance for a motor system, we hypothetically force the motor system to an equilibrium by cutting

off all chemical transitions out of the current cycle and artificially restricting the motor to $[0, L]$. The equilibrium solution is given by the Boltzmann distribution

$$\rho_j^{(e)}(x,q) \propto \exp\left(\frac{-\left(\phi_j(x)+\frac{q^2}{2m}\right)}{k_BT}\right).$$

With the Boltzmann distribution, the zero probability flux in the reaction direction leads to

$$\frac{k_{(j+1)j}(x)}{k_{j(j+1)}(x)} = \frac{\rho_{j+1}^{(e)}(x,q)}{\rho_j^{(e)}(x,q)} = \exp\left(\frac{\phi_j(x)-\phi_{j+1}(x)}{k_BT}\right), \quad 0<j<N$$

$$\frac{k_{1N}(x)}{k_{N1}(x)} = \frac{\rho_{N+1}^{(e)}(x,q)}{\rho_N^{(e)}(x,q)} = \exp\left(\frac{\phi_N(x)-\phi_1(x)+\Delta G}{k_BT}\right) \tag{7}$$

where $\Delta G \equiv \phi_{N+1}(x)-\phi_1(x) < 0$ is the free energy change in one reaction cycle. In other words, $(-\Delta G) > 0$ is the free energy released in one reaction cycle. (7) is the condition of detailed balance for transition rates $k_{ij}(x)$.

If the effect of inertia is ignored, Langevin equation (2) is approximated by

$$\zeta\frac{\mathrm{d}\mathbf{x}}{\mathrm{d}t} = -f_0 - \phi_{\mathbf{s}}'(\mathbf{x}) + \sqrt{2k_BT\zeta}\frac{\mathrm{d}\mathbf{W}(t)}{\mathrm{d}t} \tag{8}$$

and the corresponding Fokker–Planck equation is

$$\frac{\partial\rho_j(x,t)}{\partial t} = D\frac{\partial}{\partial x}\left[\frac{\phi_j'+f_0}{k_BT}\rho_j + \frac{\partial\rho_j}{\partial x}\right], \quad j=1,2,\ldots,N \tag{9}$$

where $D \equiv k_BT/\zeta$ is the diffusion coefficient.

2. Stokes efficiency, previous results and a general proof

In [11,12], the Stokes efficiency was introduced to measure how efficiently the motor uses the chemical free energy to drive through the viscous media. Let L be the period of motor potentials $\{\phi_j(x)\}$. Let $v \equiv \lim_{t\to\infty}\langle\mathbf{x}(t)\rangle/t$ be the average velocity of the motor and r the reaction rate (average number of reaction cycles per unit time) of the motor. The Stokes efficiency is defined as

$$\eta_{\text{Stokes}} \equiv \frac{\zeta v^2}{r(-\Delta G) - vf_0}. \tag{10}$$

In (10), the denominator is the net average of energy consumed per unit time: $r(-\Delta G)$ is the average free energy released from the chemical reaction per unit time and vf_0 is the average free energy output to the external agent exerting the conservative load force on the motor. The numerator ζv^2 has the dimension of energy per time. But it does not have a clear thermodynamic meaning. As a result, $\eta_{\text{Stokes}} \le 1$ cannot be derived based on simple thermodynamic arguments. In [12], for the overdamping case, Eq. (9), where the effect of inertia is ignored, we proved $\eta_{\text{Stokes}} \le 1$. Below we present a proof for the general case, Eq. (5). Specifically, we will prove $\zeta v^2 \le r(-\Delta G) - vf_0$.

First we rewrite the right hand side of Fokker–Planck equation (5) as

$$\begin{aligned}\frac{\partial\rho_j}{\partial t} &= \frac{\partial}{\partial q}\left[\left(\phi_j'\rho_j + k_BT\frac{\partial\rho_j}{\partial x}\right) + f_0\rho_j + \zeta\left(\frac{q}{m}\rho_j + k_BT\frac{\partial\rho_j}{\partial q}\right)\right] - \frac{\partial}{\partial x}\left[\frac{q}{m}\rho_j + k_BT\frac{\partial\rho_j}{\partial q}\right] + I_{j-1/2} - I_{j+1/2}\\ &= \frac{\partial}{\partial q}\left[\rho_j\frac{\partial F_j}{\partial x} + \zeta\rho_j\frac{\partial F_j}{\partial q} + \rho_j f_0\right] - \frac{\partial}{\partial x}\left[\rho_j\frac{\partial F_j}{\partial q}\right] + I_{j-1/2} - I_{j+1/2}\end{aligned} \tag{11}$$

where function $F_j(x,q,t)$ is defined as

$$F_j(x,q,t) \equiv \phi_j(x) + \frac{1}{2m}q^2 + k_BT\log(\rho_j(x,q,t)) \tag{12}$$

Thus, Eq. (5) can be written in a conservation form in terms of probability fluxes in the q-direction, in the x-direction and in the reaction direction:

$$\frac{\partial\rho_j}{\partial t} = -\frac{\partial J_j^{(q)}}{\partial q} - \frac{\partial J_j^{(x)}}{\partial x} + I_{j-1/2} - I_{j+1/2} \tag{13}$$

where fluxes $J_j^{(q)}$ and $J_j^{(x)}$ are given by

$$\begin{aligned}J_j^{(q)} &\equiv -\rho_j\left(\frac{\partial F_j}{\partial x} + \zeta\frac{\partial F_j}{\partial q} + f_0\right),\\ J_j^{(x)} &\equiv \rho_j\frac{\partial F_j}{\partial q}.\end{aligned} \tag{14}$$

The flux in the reaction direction $I_{j+1/2}$ is given in (6). The boundary conditions for (13) in the x-direction are periodic:

$$\begin{aligned}
&\phi_j(x+L) = \phi_j(x),\\
&\rho_j(x+L,q,t) = \rho_j(x,q,t),\\
&k_{(j+1)j}(x+L) = k_{(j+1)j}(x),\\
&k_{j(j+1)}(x+L) = k_{j(j+1)}(x).
\end{aligned} \tag{15}$$

The boundary conditions for (13) in the reaction direction are pseudo-periodic:

$$\begin{aligned}
&\phi_{N+j}(x) = \phi_j(x) + \Delta G,\\
&\rho_{N+j}(x,q,t) = \rho_j(x,q,t).
\end{aligned} \tag{16}$$

The average velocity and the reaction rate are expressed in terms of the steady state flux as

$$\begin{aligned}
v &= \int_0^L \int_{-\infty}^{\infty} \sum_{j=1}^{N} J_j^{(x)} \mathrm{d}q\mathrm{d}x = \int_0^L \int_{-\infty}^{\infty} \sum_{j=1}^{N} \rho_j \frac{\partial F_j}{\partial q} \mathrm{d}q\mathrm{d}x\\
r &= \int_0^L \int_{-\infty}^{\infty} I_{j+1/2} \mathrm{d}q\mathrm{d}x, \quad \text{for any } j.
\end{aligned} \tag{17}$$

Using (17) to write out ζv^2, applying the Cauchy–Schwarz inequality, and using the constraint $\int_0^L \int_{-\infty}^{\infty} \sum_{j=1}^{N} \rho_j \mathrm{d}q\mathrm{d}x = 1$, we obtain

$$\begin{aligned}
\zeta v^2 &= \zeta \left[\int_0^L \int_{-\infty}^{\infty} \sum_{j=1}^{N} \rho_j \frac{\partial F_j}{\partial q} \mathrm{d}q\mathrm{d}x \right]^2\\
&= \zeta \left[\int_0^L \int_{-\infty}^{\infty} \sum_{j=1}^{N} \sqrt{\rho_j}\sqrt{\rho_j} \frac{\partial F_j}{\partial q} \mathrm{d}q\mathrm{d}x \right]^2\\
&\le \zeta \left[\int_0^L \int_{-\infty}^{\infty} \sum_{j=1}^{N} \rho_j \mathrm{d}q\mathrm{d}x \right] \cdot \left[\int_0^L \int_{-\infty}^{\infty} \sum_{j=1}^{N} \rho_j \left(\frac{\partial F_j}{\partial q} \right)^2 \mathrm{d}q\mathrm{d}x \right]\\
&= \int_0^L \int_{-\infty}^{\infty} \sum_{j=1}^{N} \left(\zeta \rho_j \frac{\partial F_j}{\partial q} \right) \frac{\partial F_j}{\partial q} \mathrm{d}q\mathrm{d}x.
\end{aligned} \tag{18}$$

Integrating by parts in the q-direction and using (14) yields

$$\begin{aligned}
\zeta v^2 &\le -\int_0^L \int_{-\infty}^{\infty} \sum_{j=1}^{N} F_j \frac{\partial}{\partial q} \left(\zeta \rho_j \frac{\partial F_j}{\partial q} \right) \mathrm{d}q\mathrm{d}x\\
&\le \int_0^L \int_{-\infty}^{\infty} \sum_{j=1}^{N} F_j \frac{\partial}{\partial q} \left(J_j^{(q)} + \rho_j \frac{\partial F_j}{\partial x} + \rho_j f_0 \right) \mathrm{d}q\mathrm{d}x.
\end{aligned} \tag{19}$$

Using the steady state version of (13) to express $\partial J_j^{(q)}/\partial q$, we have

$$\begin{aligned}
\zeta v^2 &\le \int_0^L \int_{-\infty}^{\infty} \sum_{j=1}^{N} F_j \frac{\partial}{\partial q} \left(\rho_j \frac{\partial F_j}{\partial x} + \rho_j f_0 \right) \mathrm{d}q\mathrm{d}x - \int_0^L \int_{-\infty}^{\infty} \sum_{j=1}^{N} F_j \frac{\partial J_j^{(x)}}{\partial x} \mathrm{d}q\mathrm{d}x + \int_0^L \int_{-\infty}^{\infty} \sum_{j=1}^{N} F_j \left(I_{j-1/2} - I_{j+1/2} \right) \mathrm{d}q\mathrm{d}x\\
&\equiv T_1 + T_2 + T_3.
\end{aligned} \tag{20}$$

Applying integration by parts twice to T_1 leads to

$$\begin{aligned}
T_1 &\equiv \int_0^L \int_{-\infty}^{\infty} \sum_{j=1}^{N} F_j \frac{\partial}{\partial q} \left(\rho_j \frac{\partial F_j}{\partial x} + \rho_j f_0 \right) \mathrm{d}q\mathrm{d}x\\
&= -\int_0^L \int_{-\infty}^{\infty} \sum_{j=1}^{N} \rho_j \frac{\partial F_j}{\partial x} \frac{\partial F_j}{\partial q} \mathrm{d}q\mathrm{d}x - f_0 \int_0^L \int_{-\infty}^{\infty} \sum_{j=1}^{N} \rho_j \frac{\partial F_j}{\partial q} \mathrm{d}q\mathrm{d}x\\
&= -\int_0^L \int_{-\infty}^{\infty} \sum_{j=1}^{N} J_j^{(x)} \frac{\partial F_j}{\partial x} \mathrm{d}q\mathrm{d}x - f_0 \int_0^L \int_{-\infty}^{\infty} \sum_{j=1}^{N} J_j^{(x)} \mathrm{d}q\mathrm{d}x\\
&= \int_0^L \int_{-\infty}^{\infty} \sum_{j=1}^{N} F_j \frac{\partial J_j^{(x)}}{\partial x} \mathrm{d}q\mathrm{d}x - f_0 v.
\end{aligned} \tag{21}$$

Boundary condition (16) implies that $F_{N+j} = F_j + \Delta G$. Applying summation by parts to T_3, using $F_{N+1} = F_1 + \Delta G$ and $r = \int_0^L \int_{-\infty}^{\infty} I_{1/2} \mathrm{d}q\mathrm{d}x$, we arrive at

$$\begin{aligned} T_3 &\equiv \int_0^L \int_{-\infty}^{\infty} \sum_{j=1}^{N} F_j \left(I_{j-1/2} - I_{j+1/2}\right) \mathrm{d}q\mathrm{d}x \\ &= (-\Delta G) r + \int_0^L \int_{-\infty}^{\infty} \sum_{j=1}^{N} I_{j-1/2} \left(F_j - F_{j-1}\right) \mathrm{d}q\mathrm{d}x. \end{aligned} \tag{22}$$

In (22), the integrand has the form

$$\begin{aligned} I_{j-1/2} \left(F_j - F_{j-1}\right) &= \left(k_{j(j-1)}\rho_{j-1} - k_{(j-1)j}\rho_j\right) \cdot \left(\phi_j - \phi_{j-1} + k_B T \log \frac{\rho_j}{\rho_{j-1}}\right) \\ &= k_B T\, k_{j(j-1)} \rho_{j-1} \left(1 - \frac{k_{(j-1)j}\rho_j}{k_{j(j-1)}\rho_{j-1}}\right) \log \left[\exp\left(\frac{\phi_j - \phi_{j-1}}{k_B T}\right) \frac{\rho_j}{\rho_{j-1}}\right] \\ &= k_B T\, k_{j(j-1)} \rho_{j-1} \left(1 - Q\right) \log Q \end{aligned} \tag{23}$$

where the quantity Q is given by

$$Q = \exp\left(\frac{\phi_j - \phi_{j-1}}{k_B T}\right) \frac{\rho_j}{\rho_{j-1}} = \frac{k_{(j-1)j}\rho_j}{k_{j(j-1)}\rho_{j-1}}. \tag{24}$$

Here we have used the condition of detailed balance (7) so the two expressions for Q given in (24) are the same. Since the expression $(1 - Q) \log Q$ is always non-positive for all positive values of Q, we have $T_3 \leq (-\Delta G)\, r$. Substituting the results for T_1 and T_3 into (20), we have $\zeta v^2 \leq (-\Delta G) r - v f_0$, which leads immediately to the conclusion that

$$\eta_{\text{Stokes}} \equiv \frac{\zeta v^2}{r(-\Delta G) - v f_0} \leq 100\%.$$

In summary, for the general case where the inertia is retained in the mathematical formulation, we have proved that the Stokes efficiency is bounded by 100%.

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