

**Córdova-Figueroa and Brady Reply:** We reply to the preceding Comment [1]. In our original Letter [2] we model the motor and the reactive bath particles as (macroscopic) colloidal particles dispersed in a continuum solvent. The dynamics are governed by the Smoluchowski equation for the probability density of the  $N$ -particle configuration:  $\partial P_N/\partial t + \sum_{i=1}^N \nabla_i \cdot \mathbf{j}_i = 0$ ; where the flux of spherical particle  $i$  is  $\mathbf{j}_i = -\sum_{j=1}^N \mathbf{D}_{ij} P_N \cdot \nabla_j (\ln P_N + V_N/kT)$ , and  $V_N$  is the interaction potential. The Brownian force acting on particle  $i$  due to the random thermal fluctuations of the solvent is  $-kT \nabla_i \ln P_N$ . The diffusivity of an  $ij$  pair is  $\mathbf{D}_{ij} = kT M_{ij}$ , where  $M_{ij}$  is the hydrodynamic mobility tensor—the velocity of particle  $i$  due to a force on  $j$ . At equilibrium the Boltzmann distribution holds:  $P_N^{\text{eq}} \sim e^{-V_N/kT}$ , and  $\mathbf{j}_i = -\sum_{j=1}^N \mathbf{D}_{ij} P_N \cdot \nabla_j \ln(P_N/P_N^{\text{eq}})$ ; departures from equilibrium drive a flux.

Designate particle “1” to be the “motor” and the other  $N - 1$  particles to be “reactive” bath particles. We neglect hydrodynamic interactions implying that the mobility is that of an isolated particle  $\mathbf{D}_{ij} = (kT/6\pi\eta a_i) \mathbf{I} \delta_{ij}$ . Integrating the  $N$ -particle Smoluchowski equation over the positions of the  $N - 1$  bath particles produces the evolution equation for the motor:  $\partial P_1/\partial t + \nabla_1 \cdot \langle \mathbf{j}_1 \rangle = 0$ , where  $P_1(\mathbf{x}_1, t)$  is the probability density for finding the motor, and the average flux of the motor is given by  $\langle \mathbf{j}_1 \rangle \equiv -\mathbf{D}_{11} \cdot \nabla_1 P_1 + \sum_{j=2}^N \mathbf{D}_{1j} \cdot \int P_N \nabla_j \ln(P_N/P_N^{\text{eq}}) d\mathbf{r}_2, \dots, d\mathbf{r}_N = (-\mathbf{D}_{11} \cdot \nabla_1 \ln P_1 + \mathbf{D}_{11} \cdot \int P_{1/1} \nabla_2 \ln(P_{1/1}/P_{1/1}^{\text{eq}}) d\mathbf{r}_2) P_1$ , with  $\mathbf{r}_2 = \mathbf{x}_2 - \mathbf{x}_1$ , and we have introduced the conditional probability density for finding a bath particle relative to particle 1,  $P_{1/1}(\mathbf{r}_2|\mathbf{x}_1, t)$ , more commonly known as the pair-distribution function. For hard-sphere interactions,  $V_N = 0$  and  $P_{1/1}^{\text{eq}} = \text{const}$ , the integrand in the flux becomes  $\nabla_2 P_{1/1}$  and integration by parts gives for the average flux of particle 1:  $\langle \mathbf{j}_1 \rangle = (-\mathbf{D}_{11} \cdot \nabla_1 \ln P_1 + \mathbf{D}_{11} \cdot \oint \mathbf{n} P_{1/1} dS_{12}) P_1$ , where  $\mathbf{n}$  is the normal pointing out of particle 1 and  $S_{12}$  is the surface of contact between particles 1 and 2.

The average flux of particle 1 can be primitively written as the average particle velocity,  $\langle \mathbf{v}_1 \rangle$ , times the probability density  $P_1$ , viz.  $\langle \mathbf{j}_1 \rangle = \langle \mathbf{v}_1 \rangle P_1$ , and is composed of two terms: The first term is the normal unhindered random walk of the particle owing to Brownian motion:  $(-\mathbf{D}_{11} \cdot \nabla_1 \ln P_1) P_1 = -\mathbf{D}_{11} \cdot \nabla_1 P_1$ , where  $\mathbf{D}_{11} = kT/6\pi\eta a_1 \mathbf{I}$  is the Stokes-Einstein-Sutherland diffusivity of an isolated particle. The Brownian flux can be interpreted as a Brownian velocity,  $\mathbf{v}_1^B \equiv -\mathbf{D}_{11} \cdot \nabla_1 \ln P_1$ , times the probability density, which permits the further interpretation of a balance of forces between the hydrodynamic Stokes drag  $\mathbf{F}^{\text{hyd}} = -6\pi\eta a_1 \mathbf{v}_1^B$  and the Brownian force  $\mathbf{F}^B = -kT \nabla_1 \ln P_1$ , viz.  $\mathbf{F}^{\text{hyd}} + \mathbf{F}^B = 0$ . There is no violation of momentum conservation; there are only “internal” forces acting—the thermal fluctuations of the solvent being dissipated via Stokes drag.

The second term is the influence of the bath particles on particle 1. The probability density for finding

a bath particle relative to the motor for dilute bath particle concentrations is simply the number density of bath particles  $P_{1/1} = n$ . Thus,  $\mathbf{D}_{11} \cdot \oint \mathbf{n} P_{1/1} dS_{12} = (1/6\pi\eta a_1) \oint \mathbf{n} nkT dS_{12}$ —the integral of the “osmotic” pressure  $\Pi \equiv nkT$  over the surface of contact. In analogy to the balance of forces for the unhindered Brownian motion, the second contribution to the flux is a balance of hydrodynamic and osmotic forces, viz.  $\mathbf{F}^{\text{hyd}} + \mathbf{F}^{\text{osm}} = 0$ . Only internal forces act—just Brownian motion.

To complete the analysis one returns to the  $N$ -particle Smoluchowski equation to derive an equation for  $P_{1/1}$ , which, in general, depends on  $P_{2/1}(\mathbf{r}_2, \mathbf{r}_3|\mathbf{x}_1)$ , etc., as well as  $P_1$ ; the hierarchy was closed by diluteness. The specific equation for  $P_{1/1}$  or  $n$  depends on the problem considered. E.g., the hindering influence of the bath particles on the long-time self-diffusion of the motor. Or an initial condition with all particles located in one-half of a container—as the bath particles diffuse down their concentration gradient they entrain the motor giving the diffusive flux of one species driven by a concentration gradient of another. [At the Smoluchowski level there is no way to distinguish  $D_2O$  from  $H_2O$  (they are both solvent and/or have the same diffusivity) and thus there is no inconsistency.] Or, the bath particles could react at the surface of the motor and thereby create a variation in the local pair probability and a resulting flux of the motor, which is our model for osmotic propulsion.

We opted to omit this detailed statistical mechanical analysis and make a more intuitive connection to “osmotic” forces so that the reader would not be distracted and could focus attention on the conclusions of the work. Those interested in the details, we thought, would go to the cited references for a full treatment. One can criticize the model as perhaps not applying to the motivating experiments on catalytic nanomotors or for the neglect of hydrodynamic interactions. And the choice of the word osmotic seems to have caused confusion. But there is no violation of basic physical principles.

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[1] Frank Jülicher and Jacques Prost, preceding Comment, Phys. Rev. Lett. **103**, 079801 (2009).

[2] Ubaldo M. Córdova-Figueroa and John F. Brady, Phys. Rev. Lett. **100**, 158303 (2008).