

**Córdova-Figueroa and Brady Reply:** We would like to thank the authors of the preceding Comment [1] for taking an interest in our Letter [2], an interest sufficient to raise questions about our work. First, we never claimed to have *explained* the autonomous motion in the experiments of Paxton *et al.* (2004). Those experiments *motivated* us to think about possible mechanisms for autonomous motion and how a chemical reaction might be able to induce such motion. For sure, a number of other physical mechanisms could come into play and could be responsible for the motion seen in the experiments.

The authors object to our Letter on two grounds: (1) that it violates Newton's law of motion—action equals reaction, and (2) that it violates the first law of thermodynamics.

*Point (1): Newton's law.*—For a micron-sized particle translating at 1  $\mu\text{m}$  per second in water the Reynolds number  $\text{Re} \approx 10^{-6}$  and the acceleration of the particle is completely negligible. Newton's law reduces to  $\mathbf{F} = 0$ . The forces acting on the motor are the Stokes drag from the surrounding fluid and the “osmotic” force from the solute given by the integral of the osmotic pressure over the motor surface. At low Reynolds numbers there is no “inertial thrust”; the forces sum to zero.

When a particle of size “ $b$ ” (a reactant) collides with a particle of size “ $a$ ” (the motor) there will be equal and opposite forces and the two particles will move with their relative Stokes drag velocities given by  $v_a/v_b = -b/a$ . This is Newton's law: action equals reaction. This is precisely what is done in the Brownian dynamics (BD) simulations. Furthermore, when a reactant particle strikes the reactive portion of the motor surface we need to decide: Does it give up its (thermal)  $kT$  impulse and then react, or does it react first and therefore not give any  $kT$  kick? We performed the BD simulations both ways and the results were the same. What is important is the number of kicks (and their strength as set by the reactant and product diffusivities; see below) on each half of the motor. The chemical reaction on the motor surface establishes a concentration gradient of the reactant leading to more kicks on one side than the other causing the motor to move. Newton's law is not violated.

*Point (2): the first law of thermodynamics.*—A simple relabeling of the reactants into products, “ $b \leftrightarrow c$ ,” does not result in any net osmotic force. As we showed, what is important is  $(1 - sD_R/D_P)$ , where  $s$  is the stoichiometry of the reaction ( $R \rightarrow sP$ ), and  $D_R/D_P$  is the ratio of the reactant and product diffusivities. Only if the number of particles or their diffusivities change will there be a net osmotic force. The effect of changing the number is obvious—more particles leads to more thermal kicks. The reason the diffusivity ratio enters is that the diffusivity is the measure of the strength of the thermal kick that a reactant (or product) gives the motor.

The authors claim that since the reaction is isothermal there is no change in the internal energy of the system. And since no heat is added no work is performed  $W = \Delta U - Q = 0$ . They then conclude that since no work is performed there can be no motion of the motor.

Our motor does not violate this statement of the first law because there is no net force on the motor [see point (1) above] and therefore no work is performed—the rate of doing work is the force times the velocity. It may seem surprising that the motor can move without doing work, but this is one of the salient features of Brownian motion and can be understood from the classic example of the expansion of an ideal gas.

A partition separates two volumes,  $V_1$  and  $V_2$ . On one side are  $N$  ideal gas particles at temperature  $T$ . The partition is removed and the gas expands to fill the available volume  $V = V_1 + V_2$ . No work is done,  $W = 0$ , no heat is added  $Q = 0$ , and the internal energy does not change  $\Delta U = 0$ . The temperature of the gas also does not change ( $U = N\frac{3}{2}kT$ ). The entropy has increased by  $\Delta S = kN \ln V/V_1$ , and the free energy has decreased by  $\Delta F = -T\Delta S$ . Although no work has been done there is a net flux of material from one side to the other and there will be a net velocity associated with that flux (until the final steady state is reached). One can have motion without doing work. If a particle (call it the motor) is suspended in the gas, it will move from one side to the other as the gas expands. No work will be done, no heat will be added, but net motion of the motor will occur. The motor is simply *entrained* in the flux of the gas particles down their concentration gradient, a phenomenon known as multicomponent diffusion.

The osmotic motor uses a chemical reaction on its surface to create (autonomously) concentration gradients in the reactants and products and the motor is entrained in the entropically-driven flux of these chemical species. There is no violation of the first law of thermodynamics.

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Received 2 February 2009; published 16 April 2009

DOI: [10.1103/PhysRevLett.102.159802](https://doi.org/10.1103/PhysRevLett.102.159802)

PACS numbers: 47.57.-s, 05.40.-a, 07.10.Cm, 47.61.-k

[1] Thomas M. Fischer and Prajnaparamita Dhar, preceding Comment, Phys. Rev. Lett. **102**, 159801 (2009).

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