Supplementary Material: Clustering of self-thermophilic asymmetric dimers: the relevance of hydrodynamics

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Movies of the aggregation process

The movies showing the aggregation process with and without hydrodynamics are performed in very similar conditions. There is an ensemble of $N_s = 200$ asymmetric thermophilic dimers in a quasi-2d system with periodic boundary conditions and area fraction $\phi = 0.2$. Initially the dimers are placed on the nodes of a square lattice with random orientations, and the other specifications are the standard ones defined in the text.

S1.- Movie with the MPC-MD model (with hydrodynamic interactions)

S2.- Movie with the phoretic Brownian model (no hydrodynamic interactions)

Structure factor

From the results of the pair correlation function shown in Fig. 7, the structure factor, $S(q)$ can also be obtained. This function provides information about the ordering of a large-scale aggregate and might have the advantage of being a quantity that can be directly measured in experiments. We calculate $S(q)$ via a Fourier transform of the radial distribution function in Eq. (7)

$$S(q) = 1 + 2\pi \frac{N_s}{L^2} \int \left[g(r) - 1\right] \frac{\sin(qr)}{q} \mathrm{d}r \,, \tag{1}$$

with q the scattering vector. The structure factor is shown in Fig. S3. The $S(q)$ with and without hydrodynamic interactions are in this case very similar, resulting from the formation of a large-scale connected aggregate for both systems. The results in Fig. S3 show the displacement of the curve with hydrodynamics to smaller values of the wave vector, corresponding to the observed less compact structures and also the differences in some of the peaks, in consistency with the observations in the snapshots in Fig. 4 and the pair correlation function in Fig. 7.

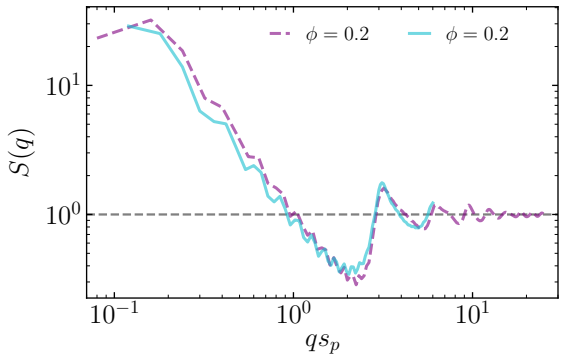

Figure S3 Structure factor between phoretic-phoretic beads for asymmetric thermophilic dimers with $\phi = 0.2$. Blue solid lines correspond to full hydrodynamic simulations (MPC+MD) and purple dashed lines to Ph-BD simulations.

Comparison with other self-phoretic dimers

In order to understand the different behaviour obtained with diffusiophoretic dimeric swimmers [1,2], we have studied the flow fields of a thermophoretic dimer with a similar geometry, namely $\gamma = 2$ philic dimers, with an extra separation between the beads of $\delta = 0.735a$. The flow field shown in Fig. S4a qualitatively resembles the one for the $\gamma = 3$ dimer with beads at contact in Fig. 1, with a different tilt in the lateral flow, which still translates into an induced torque in any neighbouring dimer. This implies that the effect of an extra separation between beads has no deep impact on the hydrodynamics. From the comparison between our results, see Fig. S4a, and the flow fields reported in Fig. 4 in Ref. [1], we already see a qualitative difference between the resulting hydrodynamics. Our flow field shows a repulsion around the head of the swimmer, which induces the hydrodynamic torque when a dimmer approaches an aggregate. The catalytic dimer shows an attractive hydrodynamic interaction on the head of the swimmer, enhancing the heads-in configuration of their clusters and, remarkably, the lateral torques are basically not present in their case. We have further studied the collective behaviour of the thermophoretic dimer showed in Fig. S4a for our typical system size and temperatures. We have obtained similar heads-out aggregation as the thermophoretic non-displaced, $\gamma = 3$ philic dimer, as shown in Fig. S4b, which are in contrast to the heads-out show by the catalytic dimers in Ref. [2]. To precisely determine the origin of the discrepancy and how to apply it to various practical cases remains a matter of further research.

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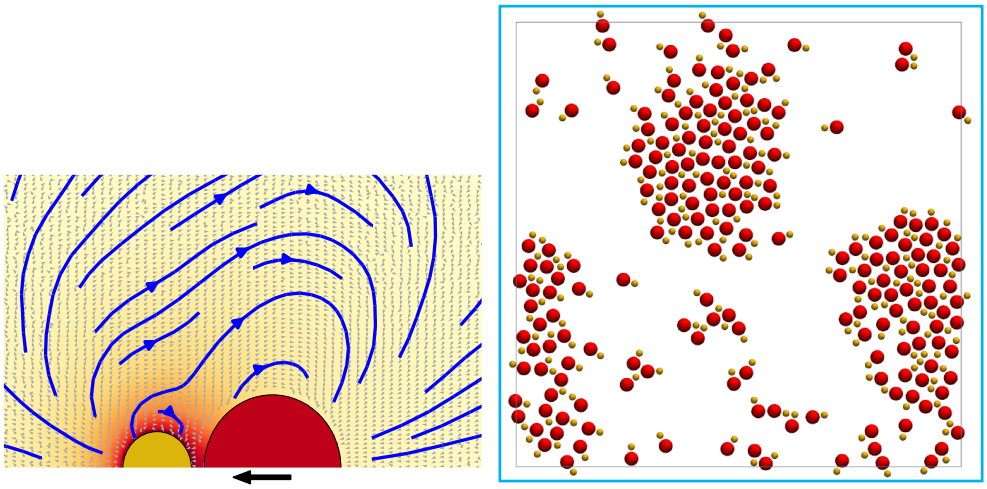

Figure S4 (a) Flow field and (b) simulation snapshot of an asymmetric $\gamma = 2$ self-thermophilic dimer with an extra bond separation δ for a time of around $320\tau_B$.

Notes and references

[1] S. Y. Reigh and R. Kapral, "Catalytic dimer nanomotors: continuum theory and microscopic dynamics," *Soft Matter*, vol. 12, pp. 3149–3158, 2015.

[2] P. H. Colberg and R. Kapral, "Many-body dynamics of chemically propelled nanomotors," *J. Chem. Phys.*, vol. 147, no. 6, p. 064910, 2017.