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Soft Matter

ARTICLE TYPE

Cite this: DOI: 00.0000/xxxxxxxxxx	Electronic supplementary information for: Active bi- nary mixtures of fast and slow hard spheres
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Received Date Accepted Date DOI: 00.0000/xxxxxxxxx	 The Electronic Supplementary Information section contains additional figures, videos and data that supplement the discussion in the main text. Movies of select data are available online. parameter_dynamics.mp4 (S3): showing several simulations initialized with different activities and fractions.
	• active_tracer.mp4 (S4): which shows tracers leaving wakes in a passive gas.

• cluster_annihilation.mp4 (S5): which shows behavior proximal to the binodal, where clusters appear briefly in time.

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S 1 Convergence study for active/active system with $Pe_s =$, $Pe_f = 500$, $x_f =$, and $\phi = 0.6$. Simulations are performed at constant area fraction and fast activity while the total number of particles (*N*) is varied to obtain different ratios of persistence length to box length ($l_p : l_{bax}$, see legend). Top row shows composition of the dense phase over time, bottom row shows the time-averaged steady-state composition of the dense phase as a function of $l_p : l_{bax}$. Columns depict results for all, slow, and fast particles respectively. At the densities and activities used in this study, ratios of persistence to box length of 1.5 maintain accurate average steady-state composition of the dense phase. Results presented in the paper for systems with $Pe_f = 500$, use $l_p : l_{bax} \ge 0.5$. Data pertaining to composition of the dense phase reaches steady state behavior rapidly ($5\tau_r$) and deviation from the average composition at long time-scales is shown to depend heavily on box size.



S 2 Remaining phase diagrams in the x_A - Pe_A plane for $Pe_B = 0 - 150$. Filled symbols denote phase-separated systems, as determined by our MIPSidentifying algorithm, and open symbols denote a gaseous steady-state. The dashed lines indicate phase boundaries predicted by the active/active equation in table 1 in the paper, which is the theoretical binodal according to our kinetic theory. Left of each plane is the same phase diagram final simulation snapshots after $100\tau_B$ (again with kinetic theory overlaid), where A particles are shown in gold and B particles in teal. Data provides good qualitative agreement with algorithmically determined phase separation as well as the kinetic theory.



S 3 (Video available online) Simulations of active Brownian particles with distinct slow (gold) and fast ($Pe_f = 500$, teal) particle fractions and activities. All snapshots are taken on the same timestep, $\tau_B = 100$. Particle fraction is constant in each row: $x_f = 0.3$, 0.5, and 0.7; illustrating majority slow, equal and majority fast mixtures respectively. Slow particle activity is constant in each column: $Pe_s = 0$, 25, 100, and 200 which is representative of the observed emergent behaviors in this space.



Initial positions

Active 'tracers' in passive bath

Magnified

S 4 (Video available online) Low fraction of fast 'tracers' ($x_f = 0.05$ and $Pe_f = 500$) in a passive bath. System does not undergo MIPS in the observed simulation time ($100\tau_B$). Fast particles push through a dense crowd of passive particles leaving trails until passive particles diffuse (or are pushed) into these voids. Magnified image depicts some examples of this effect with active particles (circled in red) and highlighted corresponding trails (transparent red tails). The ability of active particles to move through the passive bath is dependent on the local density of passive particles.



Cluster formation

Growth/upward movement

Splitting

Subcluster attrition/growth

S 5 (Video available online) Low fraction of fast particles ($x_f = 0.1$ and $Pe_f = 500$) mixed with passive particles. Motility-induced dense phases rapidly appear and dissipate. Fast particles at the cluster edge appear to push through the passive interior annihilating or dividing the clusters. While brief, clusters that form at these system parameters are highly motile (as compared to stable MIPS phases), and rapidly move across the simulation box.



S 6 Difference in composition of the dense phase (% of fast -% of slow) on a log-log scale for simulations at various constant particle fraction (top row) and slow activity (second row). Fast activity is constant for all simulations ($Pe_f = 500$). Color bars indicate activity ratio (top row) and particle fraction (second row). Increasing the net activity (via increasing Pe_s or x_f) corresponds to earlier nucleation times (crossing the x-axis). Peaks in the data appear at sufficiently high slow activity and indicate the more rapid coarsening of fast particles in the dense phase. The difference decreases in time as slow particle coarsening reaches its steady-state value. At low slow activity, slow particles do not undergo MIPS and therefore no slow particle coarsening is observed (and no peak in the percent difference). Bottom plot shows that both fast (solid red lines) and slow (dashed blue lines) particles nucleate the dense phase at the same time. This phenomenon occurs irrespective of either species activity or fraction. Nucleation shown for active passive mixture with $x_f = 0.8, 0.6, 0.4, 0.2$, left to right in nucleation times respectively.