Shear-induced assembly and breakup in suspensions of magnetic Janus particles with laterally shifted dipoles

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System Size Independence Analysis

Figure S1 illustrates the analysis for system-size independence and time independence, a qualitative validation of statistically of systems sizes (N = 125, 250, 500, 750, 1000) for simulations of s = 0.0 - 0.5 under shear flow (Pe = 1).



Fig. S1: Time-dependent aggregation of a dilute system of magnetic Janus particles at $\lambda = 45$, and Pe = 1. Panel displays the weighted-averaged mean cluster size $\langle N_c \rangle$, as a function of time for values of (s = 0.0 - 0.5) and number of particles N = 125, 250, 500, 750, 1000. Filled symbols correspond to size-independent systems, while open symbols indicate size-dependent systems

At low s < 0.2, a minimum of N=500 particles is required to ensure system size independence, sistem size N < 500 shows systemtic deviation in cluster properties of $\langle N_c \rangle$. Instead of medium $s \ge 0.2$, where systems with $N \ge 250$ particles achieve independence of system size, as evidenced by the convergent distributions of values $\langle N_c \rangle$ in Fig. S1.

To quantify the long-term stability of cluster formation, we analyzed the weighted averaged cluster size $\langle N_c \rangle$ and its fluctuations across N, s, and (Pe = 1, 5) While data collected after $t/\tau_D > 800$ suggest steady-state behavior. The data in Table S1 reveals important patterns about cluster stability that

deserve careful consideration. When we examine systems with $s \ge 0.2$, we find consistent behavior the average cluster size shows less than 1% variation for systems containing 250 or more particles. This stability suggests these conditions reliably produce predictable outcomes. However, the situation changes dramatically for s < 0.2. Here we observe much greater fluctuations, with variations reaching 7.8% for the least active systems (Pe = 1), particularly in smaller groups of particles (N < 500). systems

Table 1: Weighted Mean cluster size $\langle N_c \rangle$ and relative standard deviation (%) for systems of varying N, dipolar shift s, and Pe, evaluated at $t\tau_D > 800$

Ν	s = 0.0	s = 0.1	s0.2	s0.3	s0.4	s0.5
$t/\tau_D > 800, Pe = 1$						
125	21.907, (7.82%)	14.462, (3.36%)	8.083, (0.83%)	5.185, (0.30%)	4.268, (0.52%)	4.063, (0.16%)
250	20.707, (3.27%)	18.600, (3.03%)	8.797, (0.62%)	5.675, (0.28%)	4.432, (0.26%)	4.172, (0.17%)
500	25.888, (5.07%)	16.323, (1.60%)	9.395, (0.37%)	5.949, (0.18%)	4.574, (0.08%)	4.324, (0.17%)
750	24.945, (2.71%)	19.375, (2.03%)	8.889, (0.58%)	6.095, (0.15%)	4.584, (0.12%)	4.289, (0.11%)
1000	26.907, (3.48%)	17.942, (1.53%)	9.293, (0.31%)	6.386, (0.17%)	4.499, (0.08%)	4.821, (1.00%)
$t/\tau_D > 800, Pe = 5$						
125	13.049, (2.03%)	10.228, (1.41%)	6.531, (0.53%)	5.699, (0.35%)	4.927, (0.42%)	4.386, (0.26%)
250	12.959, (1.28%)	10.560, (0.72%)	6.859, (0.37%)	5.779, (0.26%)	5.393, (0.35%)	4.680, (0.19%)
500	14.563, (1.08%)	11.245, (0.49%)	6.857, (0.38%)	5.661, (0.19%)	5.382, (0.38%)	4.825, (0.18%)
750	13.682, (0.77%)	10.846, (0.65%)	6.860, (0.32%)	6.059, (0.32%)	5.172, (0.32%)	4.685, (0.12%)
1000	13.607, (0.72%)	12.328, (0.52%)	7.062, (0.32%)	5.854, (0.44%)	5.312, (0.12%)	4.740, (0.07%)

(Pe = 5) display approximately 50% lower variability than their passive counterparts (Pe = 1). This reduction in fluctuations may arise from shear interactions suppressing metastable configurations, though further analysis would be required to confirm this mechanism. The data also reveal that the minimum system size required for size-independent results depends critically on s: while N = 250 particles, for medium s, and low s requires $N \ge 500$ particles to achieve comparable stability. These findings provide practical guidelines for future simulations and highlight the importance of parameter-specific validation in nonequilibrium systems.

Breaking/Restoring Torque Ratio for Combined λ , s, and Pe Effects

The torque balance formalism used to analyze the behavior of magnetic Janus particles subjected to shear flow, as discussed in the subsection 3.4. To understand the quantitative nature of this competition, a torque-balance formalism is employed, building upon established methodologies used in the study of sheared Janus particles, and defining a critical ratio R_T . This ratio serves as a crucial metric for evaluating the propensity of the system to form stable aggregates versus transitioning to a dispersed, non-clustered state.

The competition between breaking torques—arising from Brownian fluctuations \mathbf{T}^B and hydrodynamic shear \mathbf{T}^H —and a restoring torque due to interparticle interaction \mathbf{T}^P has two contributions: (i) Magnetic dipole-dipole potential \mathbf{T}^P , and (ii) Dipole-dipole interactions mediated by the dipole displacement \mathbf{T}^s .

$$R_T \simeq \frac{T^{\text{Breakage}}}{T^{\text{Restoring}}} \simeq \frac{T^B + T^H}{T^P}.$$
 (1)

To quantify this competition, we defined the following characteristic magnitudes for each torque: - $\mathbf{T}^B \sim k_B T$ (Brownian torque), - $\mathbf{T}^H \sim \frac{8\pi\eta\dot{\gamma}a^3}{2}$ (hydrodynamic shear torque), - $\mathbf{T}^P \sim \mathbf{F}^{\text{Magnetic}} \sim \frac{\mu_0|\mathbf{m}|^2}{4\pi r_{d_{ij}}^4}$ (magnetic interaction torque), - $\mathbf{T}^s \sim s \cdot \frac{\mu_0|\mathbf{m}|^2}{4\pi r_{d_{ij}}^4}$ (displacement-mediated torque).

For two contacting particles with low s, the minimum distance between dipoles is $r_{d_{ij}} \sim 2a$. Due to the lateral displacement (medium s) of the dipole within each particle, the dipole-dipole separation $r_{d_{ij}}$ becomes slightly smaller than twice the radius of the particle. However, for analytical tractability and consistency in scaling arguments, we approximate $r_{d_{ij}} \sim 2a$.

To facilitate analysis, we non-dimensionalize the torques by dividing by the Brownian torque $(k_B T)$. This yields the following dimensionless groups: Pe (Péclet number) and λ (magnetic coupling parameter), as described in Section 2.1:

$$R_T \simeq \frac{1 + \frac{2}{3}Pe}{\lambda \left(1 + \frac{3}{2}s\right)}.$$
 (2)

The ratio R_T quantifies the balance between cluster aggregation and a non-clustering state in our magnetic Janus system. When $R_T \ll 1$, restorative torques predominate, facilitating stable cluster formation as dipolar alignment and strength prevail over Brownian randomness and shear-induced fragmentation. In contrast, at $R_T \gg 1$, the combined disaggregation torques compromise the magnetic interactions, resulting in the fragmentation of the cluster.

Figure S2 shows $\langle N_c \rangle$ as a function of R_T , revealing different regimes depending on the lateral displacement s, Pe, and λ . For low s, the curves collapse for $R_T > 1$. In medium s, we observe a similar trend: as λ increases, the onset of clustering shifts toward higher Pe, and s = 0.5 the region corresponding to non-clustering progressively disappears. For this dipolar shift, the threshold shows a different behavior $R_T \not\sim 1$. This suggests that for sufficiently strong magnetic interactions, the system favors chaining or aggregation regardless of the shear strength, as the restoring dipolar torque effectively suppresses disruption by Brownian and shear-induced effects.



Fig. S2: Master Curve of Combined λ and Pe Effects on Torque Ratio R_T . The panel shows the relation of $\langle N_c \rangle$ and R_T . A collapsed behavior is observed at $R_T \sim 1$ for $s \leq 0.4$.

Videos of magnetic Janus particles under shear flow

Video information:

Video V1: All The system starts with random initial positions and orientations, and have a ϕ and $\lambda = 45$ at low s = 0.0. The Random dynamics: Pe = 0 (V1_N750l45s00Pe0).

The Shear-induced dynamics: Pe = 1 (V1_N750l45s00Pe1); Pe = 20 (V1_N500l45s00Pe20)

Video V2: All The system starts with random initial positions and orientations, and have a ϕ and $\lambda = 45$ at medium s = 0.5.

The Random dynamics: Pe = 0 (V2_N500l45s05Pe0).

The Shear-induced dynamics: Pe = 1 (V2_N500l45s05Pe1); Pe = 20 (V2_N500l45s05Pe20); Pe = 300 (V2_N500l45s05Pe300)

Video V3: All The system starts with random initial positions and orientations, and have a ϕ and N = 250 at medium s = 0.5.

The Shear-induced dynamics at (Pe = 1): $\lambda = 15 \ (V4_N250l15s05Pe1), \ \lambda = 30 \ (V4_N250l30s05Pe1),$ and $\lambda = 60 \ (V4_N250l60s05Pe1),$