Supplemental Material of "Smoluchowski equations for linker-mediated irreversible aggregation"

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I. APPROXIMATION FOR THE KERNEL

Here we briefly review how a Brownian kernel is approximated by a constant kernel [1, 2]. The kernels used in this work for the two types of aggregation can be written as $K(i_1, j_1; i_2, j_2) = K_B(i_1, j_1; i_2, j_2)p(i_1, j_1; i_2, j_2)$ and $K_0(i, j) = K_{B,0}(i, j)p_0(i, j)$,, with the Brownian kernels given by

$$K_B(i_1, j_2; i_2, j_2) = 4\pi \left(D_{i_1 j_1} + D_{i_2 j_2}\right) \frac{R_{i_1 j_1} + R_{i_2 j_2}}{2}, \quad (1)$$

and

$$K_{B,0}(i, j) = 4\pi \left(D_{ij} + D_L\right) \frac{R_{ij} + R_L}{2}. \quad (2)$$

Here D_{ij}, R_{ij} are the diffusion coefficient and the size of a cluster (i, j), respectively; and D_L, R_L are the diffusion coefficient and the size of linkers, respectively. The constant kernel is obtained by assuming that $D_{ij} = D_P$ and $R_{ij} = R_P$, i.e., all clusters have the same diffusion coefficient and the same size of one single particle. Within this approximation the kernels (1,2) become,

$$K_B(i_1, j_2; i_2, j_2) = 2\kappa_P \quad (3)$$

and

$$K_{B,0}(i, j) = 2\kappa_P \alpha, \quad (4)$$

with $\kappa_P = 4\pi D_P R_P$ and

$$\alpha = \frac{1}{4}\left(1 + \frac{D_L}{D_P}\right)\left(1 + \frac{R_L}{R_P}\right). \quad (5)$$

Notice that in real systems, according to Stokes-Einstein relation, $R_L/R_P = D_P/D_L = 1/\Delta$ and thus $\alpha = (1 + \Delta)(1 + 1/\Delta)/4$.

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II. FINITE-SIZE EFFECTS

The simulations in the main text were performed on a lattice of size $N_{\rm latt} = 25^3$. In Fig. 1, the results obtained for p_2, with $\Delta = 10$ and $\phi = 0.05, 0.5$ and 0.95, on lattices of sizes $N_{\rm latt} = L^3$, with $L = 16, 25$ and 32 show that finite-size effects are negligible. All differences found are less than 1%, even in the worst case $\phi = 0.95$.

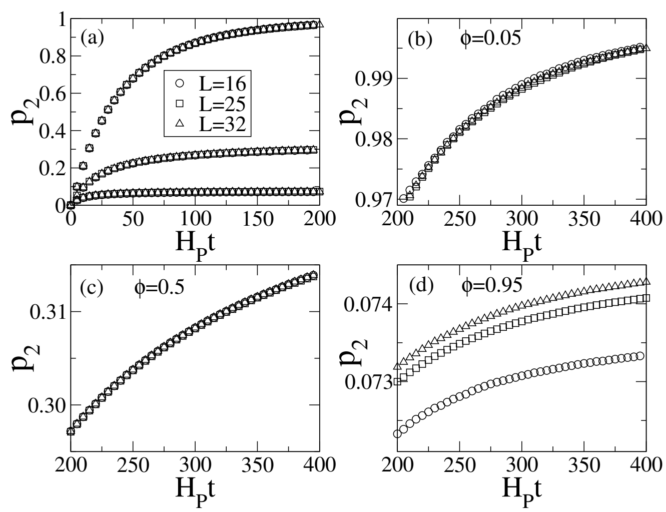

Figure 1. Analysis of finite-size effects. Results for the probability p_2 obtained with $\Delta = 10$ in simulations with $N_{\rm latt} = L^3$ sites, for $L = 16, 25$ and 32. (a) short times and $\phi = 0.05, 0.5$ and 0.95; (b) long times, $\phi = 0.05$; (c) long times, $\phi = 0.5$; (d) long times, $\phi = 0.95$.

III. ASYMPTOTIC RESULTS FOR OTHER Δ

In the main text, the asymptotic values of the probabilities for $\phi < 1 - 1/f$ are obtained with $\Delta = 10$ (simulation results). For $\Delta = 1, 5$ and 20 we obtained exactly the same asymptotic values for the probabilities: for $\phi = 0.05$, $p_2 = 1$; for $\phi = 0.5$, $p_1 = 0.6672$ and $p_2 = 0.3328$. The latter are to be compared with the results of the theory for $\phi = 0.5$: $p_2 = 1/3$ and $p_1 = 2/3$.

IV. COMPARISON TO THE CASE WITHOUT LINKERS

The results obtained in the main text can be compared to those of a system with particles (no linkers) that may bond directly to each other through their f bonding sites. In such a case, the theory with a constant kernel predicts that $p_b = \frac{2}{f}\frac{t}{1+t}$ [2]. Figure 2 compares this result to those obtained with the approximate theory for particles and linkers for several values of Δ and ϕ. At the early stages, the growth of p_b (and therefore of clusters) is much smaller when linkers are present. In the case $\phi = 0.5$, the asymptotic state is the same with or without linkers ($p_b = 2/f$). It is, however, immediately obvious that the introduction of linkers delays cluster formation even at long times. Therefore, the introduction of linkers allows for the tunable slowing down of aggregation, and in particular enables the study and exploitation of early-stage dynamics.

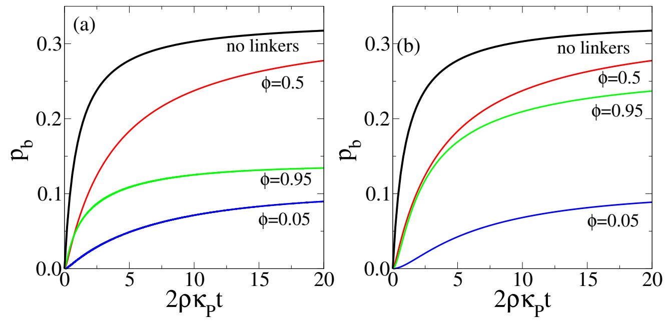

Figure 2. Bonding probability as a function of time for a system with no linkers where particles bond directly and for systems with particle-linker aggregation. (a) $\Delta = 1$, (b) $\Delta = 10$.

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[2] P. L. Krapivsky, S. Redner, and E. Ben-Naim, A Kinetic View of Statistical Physics (Cambridge University Press, New York, 2011) pp. 1–488.