Electronic Supplementary Material (ESI) for Soft Matter.

Exploiting non-equilibrium phase separation for self-assembly

Supporting Information

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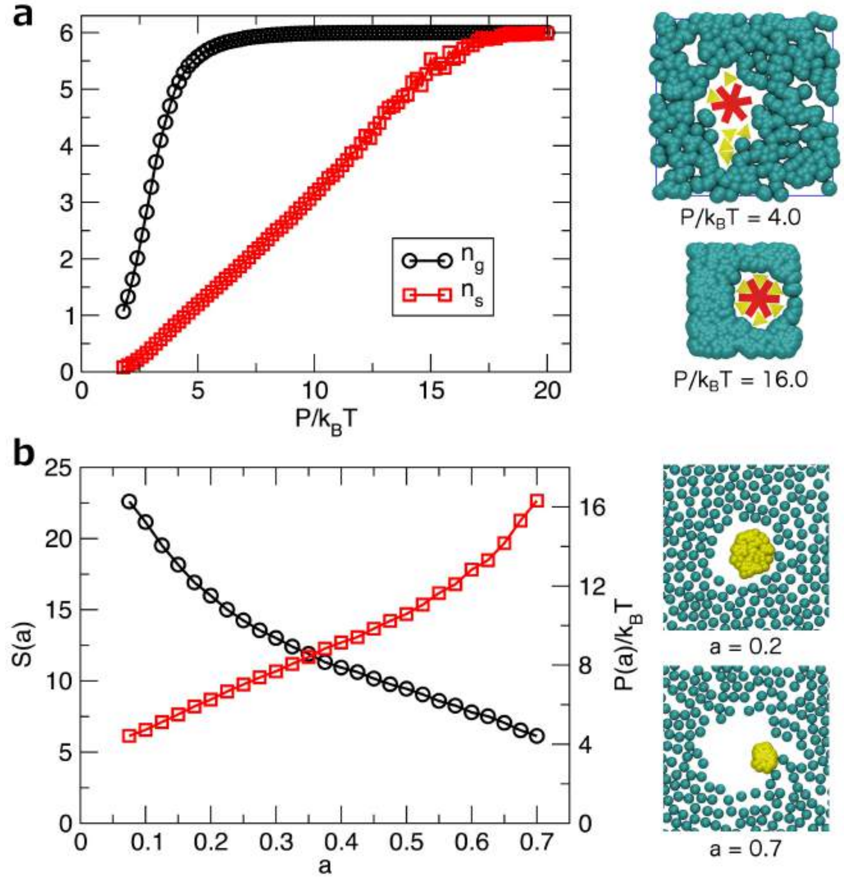
1 Correspondence between mechanical agitation and external forces

In our molecular dynamics simulations of two-dimensional systems, we use an external force $\mathbf{F}(t)$ that directly acts on particles in the simulation box. The effects of mechanical agitation on our experimental system of plastic solids is more complicated than that. Under certain assumptions, however, it can be described as an external force akin to the one used in our simulations. To establish this connection, we work in the frame of reference of the agitated board. Here, the motion of a single, isolated bead in reaction to mechanical agitation can be interpreted as originating from a force $\mathbf{G}(t)$. (Since star and wedges do not move with respect to the board unless hit by other shapes, they do not experience external forces in this frame of reference.) We can calculate $\mathbf{G}(t)$ from knowledge of the position of the board as a function of time, $\mathbf{R}(t)$. For simplicity, we assume a circular periodic motion with components $R_x(t) = R\cos(\omega t)$ and $R_y = R\sin(\omega t)$, where R is the radius of the orbital motion, and ω is the period. Assuming that beads have negligible friction with the board, the motion of a single bead in response to the agitation is $-\mathbf{R}(t)$. The corresponding force is given by $G_x(t) = mR\omega^2\cos(\omega t)$ and $G_y(t) = mR\omega^2\sin(\omega t)$, where m is the particle mass. The effects of circular periodic agitation can therefore be described as an external periodic force acting on the beads, with identical frequency and an effective amplitude of $mR\omega^2$. Note that the strength of the effective force depends on the frequency of agitation. In fact, recent experiments using a similar agitation device have shown that the tendency of shapes to demix and aggregate increases when the *frequency* of the agitation is increased [1, 2, 3]. These results agree well with the present simulations that show an increase in effective pressure and stronger effective attractions between particles with increasing *amplitude* of the external driving force.

2 Assembly of star and wedges at constant pressure

To probe the assembly behavior of the star-and-wedges at equilibrium conditions, we performed constant pressure Monte Carlo simulations with standard volume updates [4]. The simulated systems comprised the seven polyhedral shapes (with size mismatch between wedges and pockets of the star) and 439 ideal gas particles of diameter σ that serve as a pressure bath. Ideal gas particles do not have any mutual interactions but exclude area in interactions with star and wedges. These interactions favor demixing of ideal gas particles and star/wedges at high density or pressure, as illustrated in Supporting Figure 1a. Measures of the degree of aggregation and assembly increase monotonically with pressure.



SUPPORTING FIG. 1: (**a**) Average values of aggregated wedges n_{g} and assembled wedges n_{s} as a function of pressure in Monte Carlo simulations using ideal gas particles as a pressure bath. Snapshots show typical configurations at low and high pressure. (**b**) Area $S(a)$ (black circles) occupied by a cluster of 100 ideal gas particles in a bath of driven hard discs, as a function of amplitude a. Red squares indicate the corresponding effective pressure $P(a)/k_{\mathrm{B}}T$ calculated from the ideal gas equation of state. Snapshots show typical configurations at two different amplitudes. (Ideal gas particles are colored yellow.)

3 Pressure effected by phase segregation

We define the effective (surface) pressure that is exerted by a driven group of particles on an undriven, segregated group of particles by measuring the area occupied by a collection of segregated ideal gas particles. To this end, we performed Monte Carlo simulations of a system of 399 hard discs and 100 ideal gas particles. Discs undergo the biased "shaking" dynamics described in the main manuscript with $\tau = 1200$. (The size of the simulation box was chosen to match the area accessible to shaking discs to that in simulations of the star and wedges system at a packing fraction of 0.65.) For amplitudes larger than approximately $a = 0.05$, ideal gas particles form a segregated dense cluster. To measure the area S occupied by the cluster, we define a grid of square cells with edge length 1.042σ; S is defined as the total area of all cells that are occupied by at least one ideal gas particle. We then define the effective pressure using the ideal gas equation of state, $P/k_{\mathrm{B}}T = N/S$. Time averages of area $S(a)$ and pressure $P(a)$ are plotted as a function of the amplitude of shaking in Supporting Figure 1b.

References

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