I. IBI POTENTIALS: ALL DENSITIES

In Figs S1-S3 we provide the potentials optimized at each volume fraction studied in the main text ($\eta = 0.02, 0.04, 0.06$, and the additional case of $0.08$ for $n_{tgt} = 16$). Evident from these figures is the very minor role that density plays in determining the optimized potentials. Structural features largely remain fixed spatially and only a weak depression of the amplitude is found with increasing density. The larger amplitudes found at the lower densities presumably reflect the greater entropic drive the potential is competing with to release particles into the void region between clusters (i.e., greater space).

![Graph of optimized pair potentials for $n_{tgt} = 8$.](image)

FIG. S1. Optimized pair potentials for $n_{tgt} = 8$. 

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II. ADDITIONAL BOND ORIENTATIONAL ORDER ANALYSIS DATA

In Figs S4 and S5, we provide the bond-orientational order parameter distribution comparisons for $n_{tgt} = 8$ and 16, respectively. At a qualitative level, the agreement between the constrained and optimized systems is good; however, the agreement systematically decreases with decreasing cluster size as discussed in the main text.
FIG. S4. Bond-orientational order probability distributions for $n_{tgt} = 8$ at packing fraction $\eta = 0.06$. The filled grey and open blue curves indicate constrained and optimized simulations (using clusters with sizes between $n^* \pm \delta n$ as provided in the main text), respectively.

FIG. S5. Bond-orientational order probability distributions for $n_{tgt} = 16$ at packing fraction $\eta = 0.06$. The filled grey and open green curves indicate constrained and optimized simulations (using clusters with sizes between $n^* \pm \delta n$ as provided in the main text), respectively.