Buhot Reply: In the letter [1] I proposed a possible scenario for the phase-separation instability of binary mixtures of hard-core particles in the limit of high asymmetry between large and small particles relating this transition to a bond-percolation transition. In his Comment [2] A. A. Louis claims that, at least in the case of hard-spheres, the phase-separation transition is unrelated to bond-percolation. In fact, the mapping to bond-percolation relates to an instability of the homogeneous phase and, in the case of hard-spheres (or hard-disks) it gives only an upper bound for the packing fraction at the phase separation transition.

First of all, let me recall the mapping argument to the bond-percolation transition proposed in [1]. Due to the large asymmetry, the radial distribution function $g_b(r)$ of the large particles possesses a sharp and high peak of width $\sigma_s$ (size of small particles) at the contact value $\sigma_l$ between two large particles. This peak may be interpreted as "bonds" between large particles and the number of bonds $n_b$ is then defined as:

$$n_b = \rho_l \int_{\sigma_l \leq r \leq \sigma_l + \sigma_s} g_b(r) \, dr \quad (1)$$

where $\rho_l$ is the number density of the large particles. As the number $n_b$ increases with the total packing fraction, larger and larger aggregates of large particles appear in the homogeneous fluid phase. For a sufficiently large number of bonds $n_c$, or equivalently a sufficiently large packing fraction, a macroscopic aggregate appears in the system. This macroscopic aggregate breaks the translational invariance of the system which is no more homogeneous. Thus, the homogeneous fluid phase is unstable. In the letter [1] I approximate the number $n_c$ by the number of bonds $zp_c$, at the bond-percolation transition of the corresponding crystal lattice of large particles where $z$ is the coordination number and $p_c$ the bond-percolation threshold. This approximation neglects two effects: the slight modification due to the lack of lattice and, more important, the possible correlations between bonds. Taking into account those possible correlations, the appearance of the solid phase (or macroscopic aggregate) will be shift to a lower number of bonds ($n_c < zp_c$). The packing fraction $\eta_c$ corresponding to $n_b = zp_c$ is thus only an upper bound of the packing fraction $\eta_l$ at the phase-separation transition between the fluid phase and the fluid-solid phase.

In the case where the phase-separation transition is strongly first order due to a large surface tension between the fluid and the solid, we may expect large correlations between the bonds and then the bond-percolation transition is not directly related to the phase-separation transition. However, $\eta_c$ is an upper bound (and thus only a qualitative estimation) for $\eta_l$. This is exactly what is observed on Fig.1 of the Comment [2], since, for hard-spheres, we expect a large surface tension.

It is also interesting to notice that for hard-disk mixtures the prediction that instability of the homogeneous fluid phase occurs for sufficiently high asymmetry remains valid. Consequently, a phase-separation transition is predicted as already said in [1]. However, no simulations are yet able to confirm this result.

Concerning the case of parallel hard-cubes, as already said in the Comment [2], it is known that the freezing of the one component fluid is a second order transition [3]. This is principally due to the lack of rotational symmetry since the cubes are parallel [3,4] (the first order nature of the freezing transition is restored if we allow the cubes to rotate). Thus, in the case of binary mixtures of parallel hard-cubes (or squares), we may expect that the surface tension between the fluid and the solid is low. In that case, we may expect that the packing fraction $\eta_c$ is a quantitative approximation for $\eta_l$. Numerical simulations for few state points seem to confirm this result but a complete numerical calculation of the phase diagram is still lacking.

In conclusion, the phase-separation transition is not directly related to bond-percolation transition. However, the mapping proposed in [1] may be useful to predict the existence of phase separation in binary mixtures of hard-core particles since it gives an upper bound of the packing fraction at the phase separation transition. It is for example the case for the hard-disks mixture.

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