Quinn, Hong, and Luding Reply: We find it absurd that Walliser [1] essentially used the same analysis and obtained results identical to those reported in [2], yet arrived at different conclusions. Using an incomplete theory and erroneous arguments, he not only disputes the original results [3], but also claims them wrong. A more complete theory and much more detailed studies were published in [2], from which we concluded that such results support the mechanism of segregation introduced in Ref. [3]. We want to make it clear that Walliser obtained partial results of Ref. [2] and arrived at the opposite conclusion. In the following we discuss his Comment and its relevance, but at the same time point out what went wrong with his arguments.

First, the free energy functional used in Refs. [1,2] is the granular gas/fluid free energy. The observation of either the Brazil nut or the reverse Brazil nut problem [3] was, however, discussed and connected to the condensation or crystallization of the material at the bottom of the container. In such very dense situations, in the absence of convection, the geometrical size segregation can override other segregation phenomena [4]. If the system density is nowhere close to the crystallization density, the fluid free energy description is appropriate for the granular gas. The other situations, where parts of the system are condensed/crystallized, cannot be explained qualitatively by a gas/fluid free energy—as attempted in the Comment [1]—and cannot be understood by a gas/fluid based approach. The correct way of describing such crystallization is to go beyond a simple density functional approach [1,2] and use the weighted density functional approach (see Ref. [5] and references therein). Also other attempts based on Enskog theory [6], and/or empirical predictions for a global equation of state [7] and numerical modeling, at least account for the crystallization and the corresponding change of material behavior.

Such advanced methods clearly reveal the formation of crystals below the condensation temperature. (We want to point out that we used the term condensation and crystallization interchangeably.) However, the fluid free energy functional used in [1] and [2] cannot describe the formation of crystals. Hence, for the mixture of two hard spheres A and B with the condensation temperatures $T(B) < T(A)$, if the system is quenched between the two temperatures, $T(B) < T < T(A)$, the method may break down or the results are not reliable. This is why the quenching must be done from above. Nevertheless, we have considered in [2], contrary to [1], the appearance of the Brazil nut and the reverse Brazil nut problem for the system quenching $T(B) < T(A) < T$ as a positive sign to support our condensation driven segregation mechanism.

Second, according to [1], the segregation can be understood by the competition between gravity and entropy rather than condensation and percolation—this statement is based on a theory, which does not recognize condensation or percolation. Crystallization of hard spheres under gravity is due to the excluded volume interaction, and we have demonstrated analytically [6] and numerically [7], as well as with extensive molecular dynamics simulations [7,8], that such a hard sphere crystallization process does exist under gravity. Furthermore, we have extended this theory to binary mixtures in [2,3] and assumed that species are noninteracting, leading to ideal mixtures. The scenario then was tested and verified with extensive molecular dynamics simulations [3,8]. Therefore the formation of a crystallized region is well grounded, not at all controversial, and, thus, has to be accounted for. In his Comment, Walliser claims that everything can be understood using conventional thermodynamics. Without exploring all the thermodynamic aspects of the segregation, we are not prepared to dispute his argument. But here are some crucial problems with the thermodynamic argument. (i) First, consider the stability of the phase diagram obtained in [3]. For a mixture of large diameter ratio, the phase diagram [3] must break down at some point, and smaller particles on the top must percolate through the pores and sink to the bottom. It is questionable whether thermodynamics alone can describe such a time dependent stability problem. Note that thermodynamics mainly deals with the equilibrium configurations, and says nothing about the dynamical process of segregation. (ii) The correct free energy functional must survive the crystallization. Any conclusions, such as those made by Walliser [1], which are based on a pure fluid free energy functional, must be incomplete. It is very dangerous to extract conclusions from such an incomplete theory. For a single species, for example, see Ref. [5], where the weighted density functional theory does yield the crystallization near the bottom of the container.

Paul V. Quinn
Department of Physical Sciences, Kutztown University
Kutztown, Pennsylvania 19530

Daniel C. Hong
Physics, Lewis Laboratory, Lehigh University
Bethlehem, Pennsylvania 18015

Stefan Luding
Particle Technology, DelftChemTech
Julianalaan 136, 2628 BL Delft, The Netherlands

Received 14 February 2002; published 9 October 2002
DOI: 10.1103/PhysRevLett.89.189604
PACS numbers: 64.75.+g

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