Segregation in Binary Mixtures under Gravity

J. T. Jenkins and D. K. Yoon

Department of Theoretical and Applied Mechanics, Cornell University, Ithaca, New York 14853

(Received 20 November 2001; published 24 April 2002)

We employ kinetic theory for a binary mixture to study segregation by size and/or mass in a gravitational field. Simple segregation criteria are obtained for spheres and disks that are supported by numerical simulations.

DOI: 10.1103/PhysRevLett.88.194301 PACS numbers: 45.70.Mg, 05.20.Dd, 51.10.+y, 64.75.+g

Introduction.—Particle segregation in agitated systems is still not well understood, although it is important in the manufacturing of pharmaceuticals, powder metallurgy, coal and mineral processing, solid state chemistry, and geophysics. The various mechanisms of segregation that have been proposed are reviewed by Duran *et al.* [1]. They include buoyancy and preferential propping, depending on the nature of the agitation [2], and filtering by boundary layers in convection cells [3].

Recently, Hong *et al.* [4] have revisited the problem of segregation for systems in which the velocity fluctuations of the grains are nearly Maxwellian. They explore a simple criterion for segregation that is based on the idea that if one species of particle does not have sufficient kinetic energy at the bottom of a mixture to remain mobile, it will condense and segregate. However, using computer simulations, they discovered that at mass and diameter ratios at which condensation of the large spheres was predicted, the small particles could percolate through a condensed phase of large particles, forcing the large particles above them. Recognizing this competition between percolation and condensation, they proposed a relation between the diameter and the mass ratios of the two species that successfully predicted when the condensation would dominate percolation and the large particles would rise.

Here we also consider a thermalized binary mixture of grains under gravity and attempt to predict when the larger grains will rise. To do this, we adopt a simple kinetic theory for a binary mixture of spheres that differ in size and/or mass [5].

Preliminaries.—We consider spheres of two species, A and B. The spheres are assumed to be nearly elastic, smooth, and homogeneous. Spheres of species i, where i is either A or B, have radius r_i and mass m_i. We also introduce $r_{ij} \equiv r_i + r_j$ and $m_{ij} \equiv m_i + m_j$.

To define mean values, we employ the single particle velocity distribution functions $f_i^{(1)}(\mathbf{c}, \mathbf{x}, t)$, where $\mathbf{c}$ is the particle velocity, $\mathbf{x}$ is the position of the particle, and t is the time. The number density n_i of species i is, then,

$$n_i(\mathbf{x}, t) = \int f_i^{(1)}(\mathbf{c}, \mathbf{x}, t)\, d\mathbf{c},$$

where the integration is taken over all $\mathbf{c}$. The total number density n is

$$n \equiv n_A + n_B.$$

The mass density ρ_i of species i is defined as $m_i n_i$ and the total mass density ρ is

$$\rho \equiv \rho_A + \rho_B = m_A n_A + m_B n_B.$$

The mean velocity $\mathbf{u}_i$ of species i is

$$\mathbf{u}_i \equiv \langle \mathbf{c}_i \rangle = \int \mathbf{c}_i f_i^{(1)}(\mathbf{c}, \mathbf{x}, t)\, d\mathbf{c}.$$

Then, the mass average, or barycentric velocity, $\mathbf{u}$, is defined as the mass average of the species velocities:

$$\mathbf{u} \equiv \rho^{-1}(\rho_A \mathbf{u}_A + \rho_B \mathbf{u}_B).$$

We also define the diffusion velocity, $\mathbf{v}_i$, which is of central importance in this paper, because it indicates whether the motion of B relative to A is upward or downward:

$$\mathbf{v}_i \equiv \mathbf{u}_i - \mathbf{u}.$$

The temperature, T_i, of species i is defined by

$$T_i \equiv \tfrac{1}{2} m_i \langle (\mathbf{c}_i - \mathbf{u}) \cdot (\mathbf{c}_i - \mathbf{u}) \rangle,$$

which is the mean of the kinetic energy of the fluctuations relative to the barycentric velocity. The mixture temperature, T, is defined as the number average of the species temperatures:

$$T \equiv n^{-1}(n_A T_A + n_B T_B).$$

Momentum balance.—We ignore contributions to the species stress that are quadratic in the diffusion velocity and that are associated with the rate of deformation of the mixture, and we assume that the only external force is associated with the gravitational acceleration $\mathbf{g}$. In this event, the balance of momentum of species i has the form

$$\rho_i \dot{\mathbf{u}}_i = -\nabla \pi_i + n_i m_i \mathbf{g} + \boldsymbol{\phi}_i, \tag{1}$$

where the dot indicates a time derivative with respect to the mean velocity of i, π_i is the partial pressure, and $\boldsymbol{\phi}_i$ is the rate per unit volume at which momentum is provided to i in interactions with the other species. Because the interactions are equal and opposite, $\boldsymbol{\phi}_B = -\boldsymbol{\phi}_A$.

The partial pressures are given by Jenkins and Mancini [5] as

$$\pi_i = n_i T \Big(1 + \sum_{j=A,B} K_{ij} \Big),$$

where K_{ij} is defined for spheres ($D = 3$) and circular disks ($D = 2$) in terms of the species volume fraction ν_i and the radial distribution function for contacting pairs g_{ij} by

$$K_{ij} = \frac{1}{2}\,\nu_j g_{ij}\left(1 + \frac{r_i}{r_j}\right)^D .$$

For spheres, $\nu_i = 4\pi n_i r_i^3/3$ and

$$g_{ij} = \frac{1}{1-\nu} + \frac{6 r_i r_j}{r_{ij}}\frac{\xi}{(1-\nu)^2} + 8\left(\frac{r_i r_j}{r_{ij}}\right)^2 \frac{\xi^2}{(1-\nu)^3},$$

where $\nu = \nu_A + \nu_B$ is the mixture volume fraction, $\xi_i = 2\pi n_i r_i^2/3$, and $\xi = \xi_A + \xi_B$. Similarly, for circular disks, $\nu_i = \pi n_i r_i^2$ and

$$g_{ij} = \frac{1}{1-\nu} + \frac{9}{8}\frac{r_i r_j}{r_{ij}}\frac{\xi}{(1-\nu)^2},$$

where $\nu = \nu_A + \nu_B$ is the mixture area fraction, $\xi_i = \pi n_i r_i$, and $\xi = \xi_A + \xi_B$. Arnarson and Jenkins [6] provide more refined expressions for the gradients of the partial pressures that are derived in the context of the revised Enskog theory.

The interaction terms are given by Jenkins and Mancini [5] as

$$\boldsymbol{\phi}_i = K_{ij} n_i T\left[\left(\frac{m_k - m_i}{m_{ik}}\right)\nabla \ln T + \nabla \ln\left(\frac{n_i}{n_k}\right) + \frac{4}{r_{ik}}\left(\frac{2 m_i m_k}{\pi m_{ik} T}\right)^{1/2}(\mathbf{v}_k - \mathbf{v}_i)\right], \quad (2)$$

for $i \neq j$. Arnarson and Jenkins [6] show that this expression for the interactions differs from that of the revised Enskog theory by a term associated with a thermal diffusion coefficient. However, in a dense system, this term is negligible compared to the term involving ∇T that appears in (2). The quantity $\mathbf{v}_k - \mathbf{v}_i$ is the relative motion of the two species; here we are interested in the sign of the vertical component of this velocity difference.

When the momentum balances are weighted by the inverse of the densities and subtracted, the only inertia terms that survive are associated with the diffusion velocity, and we neglect them. Then, the weighted difference is

$$\mathbf{0} = -\frac{1}{\rho_A}\nabla\pi_A + \frac{1}{\rho_B}\nabla\pi_B + \frac{1}{\rho_A}\boldsymbol{\phi}_A - \frac{1}{\rho_B}\boldsymbol{\phi}_B$$

or, with $\boldsymbol{\phi}_B = -\boldsymbol{\phi}_A$,

$$\mathbf{0} = -\rho_B \nabla\pi_A + \rho_A \nabla\pi_B + \rho\boldsymbol{\phi}_A . \quad (3)$$

The segregation criterion.—We employ Eqs. (1) and (3) to describe the state of a general binary mixture under gravity, ignore the mixture inertia, and attempt to determine in what direction the segregation of species A and B will occur. After we do this, we consider the special case in which the large particles are dilute in a dense gas of small particles. That is, $r_B > r_A$ and $n_B/n_A \ll 1$.

We suppose that all gradients are parallel to gravity, in which case (3) may be written as

$$0 = -\frac{\rho_B}{\rho}\pi_A' + \frac{\rho_A}{\rho}\pi_B' + \phi_A , \quad (4)$$

where the prime indicates a derivative with respect to the vertical coordinate. Similarly, upon ignoring the inertia term in the mixture Eq. (1) becomes

$$\pi_A' = -\rho_A g + \phi_A . \quad (5)$$

Next, we introduce R, the weighted ratio of the partial pressures,

$$R \equiv \frac{\pi_A}{\pi_B}\frac{n_B}{n_A}. \quad (6)$$

We note that R is dependent on ν_i, g_{ij}, and r_i/r_j. Then

$$\begin{aligned}\pi_B' &= \left(\frac{\pi_A}{R}\frac{n_B}{n_A}\right)' \\ &= \left(\frac{n_B}{n_A}\right)'\frac{\pi_A}{R} + \frac{n_B}{n_A}\frac{\pi_A'}{R} - \frac{n_B}{n_A}\frac{\pi_A}{R}\frac{R'}{R},\end{aligned}$$

or, upon employing (5) and (6),

$$\pi_B' = \pi_B\left[\ln\left(\frac{n_B}{n_A}\right)\right]' - \frac{m_A n_B g}{R} + \frac{n_B}{n_A}\frac{\phi_A}{R} - \pi_B(\ln R)'. \quad (7)$$

We employ the expressions (5) and (7) for the gradients of the partial pressures π_A and π_B in (4) and, after some simplification, obtain the relation

$$\begin{aligned}0 = {}& \pi_B\left(\ln R - \ln\frac{n_B}{n_A}\right)' + g\,\frac{n_B}{R}(m_A - R m_B) \\ & - \left(1 + \frac{n_B}{n_A}\frac{1}{R}\right)\phi_A ,\end{aligned}$$

where

$$\begin{aligned}\phi_A = K_{AB} n_A T\Bigg\{&\left(\frac{m_B - m_A}{m_{AB}}\right)(\ln T)' + \left[\ln\left(\frac{n_B}{n_A}\right)\right]' \\ &+ \frac{4}{r_{AB}}\left(\frac{2 m_A m_B}{\pi m_{AB} T}\right)^{1/2}(v_B - v_A)\Bigg\}.\end{aligned}$$

Finally, we introduce the vertical component of the relative diffusion velocity $w_{BA} = v_B - v_A$ and suppose that the temperature is uniform. Then

$$K_{AB}\left\{\frac{4}{r_{AB}}\left(\frac{2 m_A m_B}{\pi m_{AB} T}\right)^{1/2} w_{BA} + \left[\ln\left(\frac{n_B}{n_A}\right)\right]'\right\} = \frac{1}{n_A + R^{-1} n_B}\left[\pi_B(\ln R)' + \frac{n_B}{R}(m_A - R m_B)g\right]. \quad (8)$$

In Eq. (8), the influence on the initial segregation of the differences in mass and size has been made explicit.

We note that even in the absence of a temperature gradient, segregation is possible. Here we explore this further, by neglecting the inhomogeneities in both the temperature and the mixture volume fraction. It is possible to show that $(\ln R)'$ is small in comparison to the term that remains. On ignoring this term, we obtain a very simple equation relating the relative velocity to the ratio of the mass of each species and the ratio of partial pressures:

$$\frac{4K_{AB}T}{r_{AB}}\left(\frac{2m_Am_B}{\pi m_{AB}T}\right)^{1/2} w_{BA} = \frac{n_B}{Rn_A + n_B}(m_A - Rm_B)g\,. \tag{9}$$

If $m_A > Rm_B$, then $w_{BA} > 0$ and species B will rise with respect to A. On the other hand, if $m_A < Rm_B$, $w_{BA} < 0$, and B will fall with respect to A.

Special case: $n_B/n_A \ll 1$ for spheres.—As promised, we consider the case when the larger diameter spheres B are dilute and smaller diameter spheres A are dense. In this case, $n_B/n_A \ll 1$ and several approximations can be made:

$$g_{AA} \doteq \frac{1}{1-\nu} + \frac{3\nu}{2(1-\nu)^2} + \frac{\nu^2}{2(1-\nu)^3}\,,$$

$$g_{AB} \doteq \frac{1}{1-\nu} + \frac{3\nu}{(\eta+1)(1-\nu)^2} + \frac{2\nu^2}{(\eta+1)^2(1-\nu)^3}\,,$$

and

$$\begin{aligned} R &\equiv \frac{\pi_A}{\pi_B}\frac{n_B}{n_A} \doteq \frac{g_{AA}}{g_{AB}}\left(\frac{r_{AA}}{r_{AB}}\right)^3 \\ &= \left(\frac{2\eta}{1+\eta}\right)^3 \\ &\quad\times \frac{(\eta+1)^2[2(1-\nu)^2 + 3\nu(1-\nu) + \nu^2]}{2[(1-\nu)^2(\eta+1)^2 + 3\nu(1-\nu)(\eta+1) + 2\nu^2]}\,, \end{aligned}$$

where $\eta \equiv r_A/r_B$. We express this condition more simply by evaluating R at $\nu = 1/2$. In this case,

$$R = \frac{24\eta^3}{(\eta+1)(\eta+2)(\eta+3)}\,.$$

Then, when μ is defined as the ratio of the density of the material of sphere of B to that of sphere A,

$$\mu \equiv \frac{m_B r_A^3}{m_A r_B^3} = \frac{m_B}{m_A}\eta^3,$$

the sign of w_{BA} is, by (9), the same as that of

$$(\eta+1)(\eta+2)(\eta+3) - 24\mu\,.$$

In Fig. 1 we plot $\eta \equiv r_A/r_B$ versus $\mu \equiv \eta^3 m_B/m_A$ and indicate on it the curve $\mu = (\eta+1)(\eta+2)(\eta+3)/24$ (solid line). For the values of μ above this curve, the large spheres rise; for values of μ below it, they fall. Also shown is the criterion $\mu = \eta$ of Hong *et al.* [4] based on a competition between condensation and percolation (dashed line). Their numerical simulations provide support for this criterion and, because the two curves in Fig. 1 are relatively close, they also provide support for the present criterion. The value 1/2 for the mixture volume fraction was chosen because it is a simple ratio at a representative dense state. No attempt was made to fit the data of Hong *et al.*

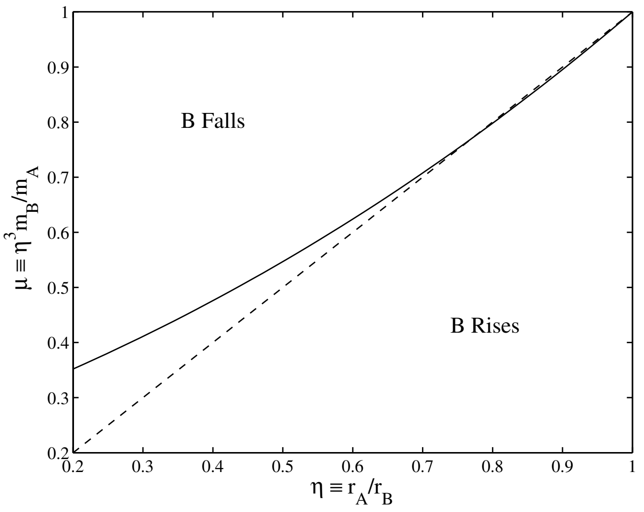

FIG. 1. Density ratio versus radii ratio for spheres.

Special case: $n_B/n_A \ll 1$ for disks.—As in the case of spheres, we make the following approximations:

$$g_{AA} \doteq \frac{1}{1-\nu} + \frac{9}{16}\frac{\nu}{(1-\nu)^2}\,,$$

$$g_{AB} \doteq \frac{1}{1-\nu} + \frac{9}{8}\frac{\nu}{(\eta+1)(1-\nu)^2}\,,$$

and

$$\begin{aligned} R &\equiv \frac{\pi_A}{\pi_B}\frac{n_B}{n_A} \doteq \frac{g_{AA}}{g_{AB}}\left(\frac{r_{AA}}{r_{AB}}\right)^2 \\ &= \left(\frac{2\eta}{\eta+1}\right)^2\frac{\eta+1}{2}\,\frac{16(1-\nu)+9\nu}{8(\eta+1)(1-\nu)+9\nu}\,, \end{aligned}$$

where, again, $\eta \equiv r_A/r_B$.

In this case, with $\nu = 3/4$ as a simple ratio at a representative dense state,

$$R = \frac{86\eta^2}{(\eta+1)(8\eta+35)}\,.$$

Then, upon introducing

$$\mu \equiv \frac{m_B r_B^2}{m_A r_A^2}\,,$$

we can write

$$m_A - Rm_B = 0$$

as

$$86\mu = (\eta+1)(8\eta+35)\,.$$

Similarly, as in the case of spheres, we plot η versus μ for disks in Fig. 2 and indicate the curve $\mu = (\eta+1)(8\eta+35)/86$ (solid line). We also give the criterion $\mu = \eta$ of Hong *et al.* [4] (dashed line) and note the similar shapes of

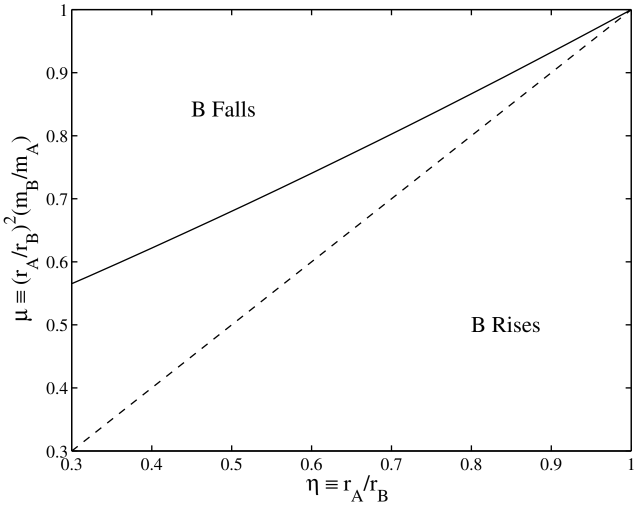

FIG. 2. Density ratio versus radii ratio for disks.

the two curves, though the case of spheres gives a closer result.

Discussion.—Our criteria for segregation are relatively close to those of Hong *et al.* [4], even though the proposed mechanisms are very different. The mechanism of Hong *et al.* depends upon there being a range of temperature over which the entire depth of one species is not completely fluidized in the presence of gravity. Also, Hong *et al.* neglect interactions between the two species, while interactions are crucial to our mechanism. When gradients are neglected, segregation in the kinetic theory is due to a competition between the inertia of the particles, through the ratio of their masses, and their geometry, as it enters through the dense corrections to the partial pressures. Because there is no relation between this segregation and the displaced volume of one or the other of the species, we do not associate this with buoyancy. Finally, we note that when both species are dilute, such segregation is predicted for particles that differ in mass, no matter what their radii may be.

We are grateful to Hans Herrmann and Stefan Luding for conversations related to particle segregation. This work was supported by the NASA Microgravity Grants No. NCC3-797 and No. NAG3-2353.

[1] J. Duran, J. Rajchenbach, and E. Clément, Phys. Rev. Lett. **70**, 2431 (1993).
[2] A. Rosato, K. J. Strandburg, F. Prinz, and R. H. Swendsen, Phys. Rev. Lett. **58**, 1038 (1987).
[3] J. B. Knight, E. E. Ehrich, V. Y. Kuperman, J. K. Flint, H. M. Jaeger, and S. R. Nagel, Phys. Rev. E **54**, 5726 (1996).
[4] D. C. Hong, P. V. Quinn, and S. Luding, Phys. Rev. Lett. **86**, 3423 (2001).
[5] J. T. Jenkins and F. Mancini, J. Appl. Mech. **109**, 27 (1987).
[6] B. O. Arnarson and J. T. Jenkins, in *Traffic and Granular Flow '99,* edited by D. Helbing, H. J. Herrmann, M. Schreckenberg, and D. E. Wolf (Springer, Berlin, 2000), pp. 481–487.