EUROPHYSICS LETTERS

OFFPRINT

Vol. 75 • Number 4 • pp. 521–527

Segregation in granular binary mixtures: Thermal diffusion

* * *

V. Garzó

Published under the scientific responsibility of the
EUROPEAN PHYSICAL SOCIETY

Incorporating
JOURNAL DE PHYSIQUE LETTRES • LETTERE AL NUOVO CIMENTO

EDP SCIENCES

EUROPHYSICS LETTERS

Europhysics Letter was launched more than fifteen years ago by the European Physical Society, the Société Française de Physique, the Società Italiana di Fisica and the Institute of Physics (UK) and owned now by 17 National Physical Societies/Institutes.

Europhysics Letters aims to publish short papers containing non-trivial new results, ideas, concepts, experimental methods, theoretical treatments, etc. which are of broad interest and importance to one or several sections of the physics community.

Europhysics letters provides a platform for scientists from all over the world to offer their results to an international readership.

EUROPHYSICS LETTERS 15 August 2006

Europhys. Lett., **75** (4), pp. 521–527 (2006)
DOI: 10.1209/epl/i2006-10143-4

Segregation in granular binary mixtures: Thermal diffusion

V. Garzó(*)
Departamento de Física, Universidad de Extremadura - E-06071 Badajoz, Spain

received 13 March 2006; accepted in final form 21 June 2006
published online 7 July 2006

PACS. 05.20.Dd – Kinetic theory.
PACS. 45.70.Mg – Granular flow: mixing, segregation and stratification.
PACS. 51.10.+y – Kinetic and transport theory of gases.

Abstract. – A solution of the inelastic Boltzmann equation that applies for strong dissipation and takes into account non-equipartition of energy is used to derive an explicit expression for the thermal diffusion factor. This parameter provides a criterion for segregation that involves all the parameters of the granular binary mixture (composition, masses, sizes, and coefficients of restitution). The present work is consistent with recent experimental results and extends previous results obtained in the intruder limit case.

Segregation and mixing of dissimilar grains is perhaps one of the most interesting problems in agitated granular mixtures. In some processes it is a desired and useful effect to separate particles of different types, while in other situations it is undesired and can be difficult to control. Several mechanisms for segregation corresponding to different scenarios have been proposed [1], but the problem is not completely understood yet. Among the different competing mechanisms, thermal diffusion becomes one of the most relevant mechanisms at large shaking amplitude where the sample of macroscopic grains resembles a granular gas. In this regime, binary collisions prevail and kinetic theory can be quite useful to analyze the physical mechanisms involved in segregation processes. Very recently, Schröter *et al.* [2] have carried out experiments in agitated mixtures constituted by particles of the same density. To the best of my knowledge, this is one of the few experiments in which thermal diffusion has been isolated from the remaining segregation mechanisms. When convection is practically suppressed, they report results to assess the influence of composition and dissipation on thermal diffusion. Given that this effect can be predicted by kinetic theory, in this letter I study the particle segregation problem driven by the presence of a thermal gradient in order to explain some of the trends observed in the above experiments [2] at large shaking amplitudes. This is the main motivation of this letter.

Under the above conditions, the so-called thermal diffusion factor Λ_{ij} characterizes the amount of segregation parallel to the temperature gradient. Thermal diffusion is caused by the relative motion of the components of a mixture due to the presence of a temperature gradient. Due to this motion, concentration gradients subsequently appear in the mixture

(*) E-mail: vicenteg@unex.es

producing diffusion that tends to oppose those gradients. A steady state is finally achieved in which the separation effect arising from thermal diffusion is compensated by the diffusion effect. Here, I determine Λ_{ij} from a solution [3] of the inelastic Boltzmann equation that applies for strong dissipation and takes into account non-equipartition of energy. This latter feature, not present in the elastic case at equilibrium, has significant consequences on the transport coefficients of the granular mixture [3].

The model system considered is a low-density binary mixture composed by smooth inelastic hard spheres ($d = 3$) or disks ($d = 2$) of masses m_i and diameters σ_i. Without loss of generality, we assume that $\sigma_1 > \sigma_2$. The inelasticity of collisions among all pairs is characterized by three independent constant coefficients of restitution α_{11}, α_{22}, and $\alpha_{12} = \alpha_{21}$. The mixture is in presence of the gravitational field $\boldsymbol{g} = -g\hat{\boldsymbol{e}}_z$, where g is a positive constant and $\hat{\boldsymbol{e}}_z$ is the unit vector in the positive direction of the z-axis. In experiments [2], the energy is usually supplied by vibrating horizontal walls so that the system reaches a steady state whose properties are presumed to be, far from the walls, insensitive to the details of the driving forces. Due to the technical difficulties involved in incorporating oscillating boundary conditions, here particles are assumed to be heated by the action of a stochastic-driving force which mimics a thermal bath. Although the relation between this driven idealized method with the use of locally driven wall forces is not completely understood, it must be remarked that in the case of boundary conditions corresponding to a sawtooth vibration of one wall the condition to determine the temperature ratio coincides with the one derived from the stochastic force [4]. This suggests that the results obtained from this driving method are a plausible first approximation for qualitative comparisons with experimental results. As will be shown below, the results derived here for the temperature ratio confirm this expectation.

The thermal diffusion factor Λ_{ij} ($i \neq j$) is defined at the steady state in which the mass fluxes $\boldsymbol{j}_i$ vanish. Under these conditions, the factor Λ_{ij} is given through the relation

$$-\Lambda_{ij}\nabla \ln T = \frac{1}{x_i x_j}\nabla x_i, \qquad \Lambda_{ij} + \Lambda_{ji} = 0, \tag{1}$$

where $x_i = n_i/n$ is the mole fraction of species i (n_i is the number density of species i and $n = n_1 + n_2$). The physical meaning of Λ_{ij} can be described by considering a granular binary mixture held between plates at different temperatures T and T' under gravity. For the sake of concreteness, we will assume that gravity and thermal gradient point in parallel directions, *i.e.*, the bottom is hotter than the top. In the steady state ($\boldsymbol{j}_1 = \boldsymbol{j}_2 = 0$), eq. (1) describes how the thermal field is related to the composition of the mixture. Assuming that Λ_{12} is constant over the relevant ranges of temperature and composition, integration of eq. (1) yields $\ln(x_1 x_2'/x_2 x_1') = \Lambda_{12}\ln(T'/T)$. Here, x_i refers to the mole fraction of species i at the plate with temperature T and x_i' refers to the mole fraction of species i at the plate with temperature T'. Consequently, if $T' > T$ and $\Lambda_{12} > 0$, then $x_1' < x_1$ while if $\Lambda_{12} < 0$, then $x_1' > x_1$. In summary, when $\Lambda_{12} > 0$, the larger particles accumulate at the top of the sample (cold plate), while if $\Lambda_{12} < 0$, the larger particles accumulate at the bottom of the sample (hot plate). The former situation is referred to as the Brazil-nut effect (BNE) while the latter is called the reverse Brazil-nut effect (RBNE).

The RBNE was observed first by Hong *et al.* [5] in MD simulations of vertically vibrated systems. They proposed a very simple segregation criterion that was later confirmed by Jenkins and Yoon [6] by using kinetic theory. More recently, Breu *et al.* [7] have experimentally investigated conditions under which the large particles sink to the bottom and claim that their experiments confirm the theory of Hong *et al.* [5] provided a number of conditions are chosen carefully. In addition to the vertically vibrated systems, some works have also focussed in the

last few years on horizontally driven systems showing some similarities to the BNE and its reverse form [8]. However, it is important to note that the criterion given in ref. [5] is based on some key assumptions: particles are assumed to be elastic, homogenous temperature and energy equipartition. These conditions preclude a comparison of the present theory with the above simulations. Previous theoretical attempts to assess the influence of non-equipartition on segregation have been recently published. Thus, Trujillo *et al.* [9] have derived an evolution equation for the relative velocity of the intruders starting from the kinetic theory proposed by Jenkins and Yoon [6] that applies for weak dissipation. They use constitutive relations for partial pressures that take into account the breakdown of energy equipartition between the two species. However, the influence of temperature gradients which exist in the vibro-fluidized regime is neglected in ref. [9] because they assume that the pressure and temperature are constant in the absence of the intruder. A more refined theory has been recently provided by Brey *et al.* [10] in the case of a single intruder in a vibrated granular mixture under gravity. The present work covers some of the aspects not accounted for in the previous theories [6,9,10] since it is based on a kinetic theory [3] that goes beyond the quasi-elastic limit [6,9] and applies for arbitrary composition x_1 (and so, it reduces to the results obtained in ref. [10] when $x_1 \to 0$). This allows one to assess the influence of composition and dissipation on thermal diffusion in bi-disperse granular gases without any restriction on the parameter space of the system. This the main value added of this paper and can be relevant to make comparisons with experiments/simulations in the dilute regime.

To determine the dependence of the coefficient Λ_{12} on the parameters of the system, we focus our attention on an *inhomogeneous* steady state with zero mass flux and gradients only along the vertical direction (z-axis). Since the flow velocity vanishes, the momentum balance equation yields $\partial_z p = -\rho g$, where $p = nT$ is the pressure, $\rho = \rho_1 + \rho_2$, and $\rho_i = m_i n_i$ is the mass density of species i. To first order in the gradients, the constitutive equation for the mass flux $j_{1,z}$ is

$$j_{1,z} = -\frac{m_1 m_2 n}{\rho} D \partial_z x_1 - \frac{\rho}{p} D_p \partial_z p - \frac{\rho}{T} D' \partial_z T, \tag{2}$$

where D is the mutual diffusion coefficient, D_p is the pressure diffusion coefficient, and D' is the thermal diffusion coefficient. The condition $j_{1,z} = 0$ yields $\partial_z x_1 = (\rho^3/m_1 m_2 n p)(D_p/D) g - (\rho^2/m_1 m_2 p)(D'/D)\partial_z T$. Substitution of this relation into eq. (1) leads to

$$\Lambda_{12} = \frac{n\rho^2}{\rho_1 \rho_2} \frac{D' - D_p g^*}{D}, \tag{3}$$

where $g^* \equiv \rho g / n \partial_z T < 0$ is the reduced gravity acceleration.

Since the mutual diffusion coefficient D is positive [3], the sign of Λ_{12} is determined by the sign of the quantity $D' - D_p g^*$. Explicit expressions for the coefficients D' and D_p have been recently obtained from the Chapman-Enskog method in the first Sonine approximation [3]. Given that the driving-stochastic term does not play a neutral role in the transport, it must be remarked that the expressions for the transport coefficients obtained in the driven case slightly differs from the ones derived in the free cooling case. In particular, while in the former situation the coefficient $D' = 0$, the thermal diffusion coefficient $D' = -(\zeta/2\nu) D_p$ in the free case [3]. Since the collision frequency ν and the total cooling rate ζ are positive, then $\mathrm{sgn}(D') = \mathrm{sgn}(-D_p)$. Consequently, the sign of Λ_{12} is the same as that of the pressure diffusion coefficient D_p in the driving-stochastic case while $\mathrm{sgn}(\Lambda_{12}) = \mathrm{sgn}(-D_p[(\zeta/2\nu) + g^*])$ in the free case.

Because of $\mathrm{sgn}(\Lambda_{12}) = \mathrm{sgn}(D_p)$ in the driven case, we will focussed henceforth on the

explicit dependence of D_p on the parameters of the system. Its expression is [3]

$$D_p = \frac{\rho_1 p}{\rho^2 \nu} \frac{x_2}{x_2 + x_1 \gamma} \left(\frac{\gamma}{\mu} - 1 \right), \tag{4}$$

where $\mu = m_1/m_2$ is the mass ratio, $\gamma = T_1/T_2$ is the temperature ratio, and ν is an effective collision frequency [3].

The condition $\Lambda_{12} = 0$ (or equivalently, $D_p = 0$ in the driven case) provides the criterion for the transition from BNE to RBNE. Equation (4) shows that the sign of D_p is determined by the value of the control parameter $\theta \equiv \gamma/\mu$. This parameter gives the mean-square velocity of the large particles relative to that of the small particles. Thus, when $\theta > 1$ ($\theta < 1$), the thermal diffusion factor is positive (negative) which leads to BNE (RBNE). The criterion for the transition condition from BNE to RBNE is $\gamma = \mu$. In the case of equal granular temperatures (energy equipartition), $\theta \to \mu^{-1}$ and so, segregation is predicted for particles that differ in mass, no matter what their diameters may be. It must be remarked that, due to the lack of energy equipartition, the condition $\theta = 1$ is rather complicated since it involves all the parameter space of the system. In particular, even when the species differ only by their respective coefficients of restitution they also segregate when subject to a temperature gradient. It is a novel pure effect of inelasticity on segregation. The criterion for the transition BNE $\Longleftrightarrow$ RBNE is the same as the one found previously in ref. [9] when α_{ij} is close to 1 and in ref. [10] in the intruder limit case ($x_1 \to 0$). However, as said before, the results obtained here are more general since they cover all the range of the parameter space of the system.

To get the explicit dependence of the control parameter θ on α_{ij}, one still needs to compute the temperature ratio γ. When the system is driven by means of a stochastic thermostat, the temperature ratio is determined from the condition [4]

$$\gamma \zeta_1 = \mu \zeta_2 \quad \text{(white-noise thermostat)}, \tag{5}$$

where the explicit form of ζ_i can be found in ref. [3]. The condition (5) differs from the one derived in the undriven (free cooling) case [11], where γ is obtained by requiring that the partial cooling rates ζ_i must be equal, *i.e.*,

$$\zeta_1 = \zeta_2 \quad \text{(free cooling)}. \tag{6}$$

This latter condition was used in ref. [10] to determine the difference of temperatures between an impurity and the surrounding gas in an open vibrated granular system.

As said above, experiments and molecular dynamics (MD) simulations have been carried out very recently [2] to analyze segregation in agitated binary granular mixtures composed by spheres of the same material and therefore, the same mass density (*i.e.*, $\mu = (\sigma_1/\sigma_2)^3$) and mechanical properties ($\alpha_{ij} = \alpha$). Figure 1 shows the comparison of the temperature ratio T_1/T_2 between MD simulations [2] and kinetic theory results based on the conditions (5) (driven case) and (6) (undriven case). The experimental value of the coefficient of normal restitution is $\alpha = 0.78$ and equal volumes of large and small particles are taken, *i.e.*, $x_2 = (\sigma_1/\sigma_2)^3 x_1$. While a good agreement between kinetic theory and MD simulations is found when the gas is assumed to be driven by a stochastic thermostat, significant discrepancies appear in the undriven case, especially as the size ratio σ_1/σ_2 increases. These results contrast with the ones obtained in ref. [10] in the tracer limit ($x_1 \to 0$) where the predictions of T_1/T_2 from kinetic theory based on the condition $\zeta_1 = \zeta_2$ compare quite well with MD simulations. However, it must noticed that for the cases studied in ref. [10] the conditions (5) and (6) yield quite similar results for the dependence of the temperature ratio on the parameters of the system.

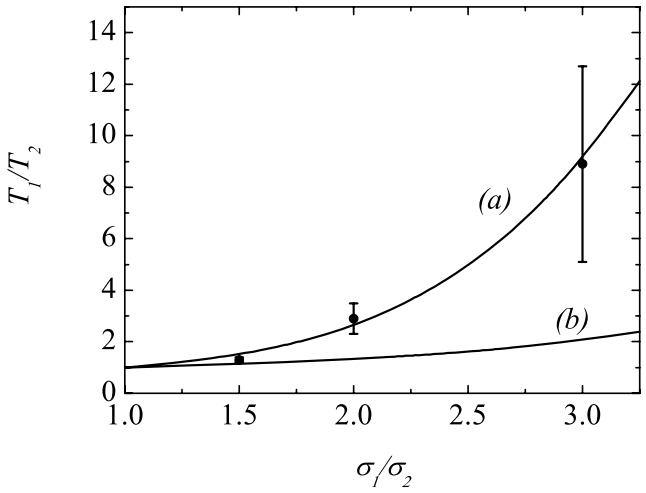

Fig. 1 – Temperature ratio T_1/T_2 *vs.* the size ratio σ_1/σ_2 for $\alpha_{ij} \equiv \alpha = 0.78$ in the case of mixtures constituted by particles of the same mass density and equal total volumes of large and small particles. The lines are the kinetic theory results in (a) the stochastic-driving case and (b) the free cooling case while the points refer to MD simulations [2].

Consider next size segregation driven by thermal difusion. To make some contact with the recent experimental results of Schröter *et al.* [2], let us consider again three-dimensional ($d = 3$) mixtures constituted by particles with the same mass density and equal total volumes of large and small particles. Figure 2 shows the phase diagram for this kind of systems. The results show that, for a given value of the coefficient of restitution, the RBNE is dominant at

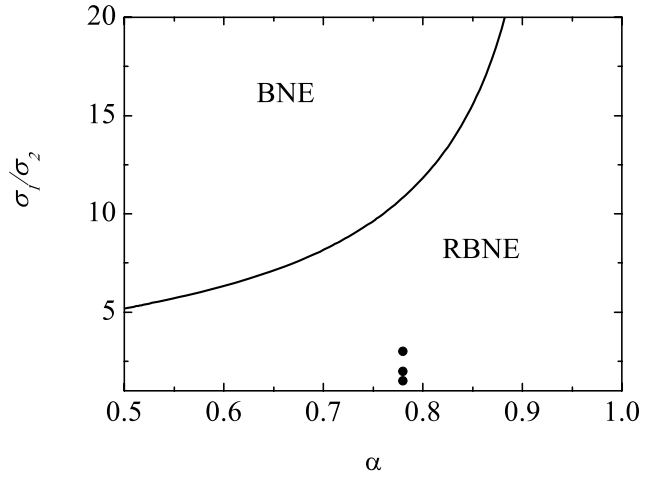

Fig. 2 – Phase diagram for BNE/RBNE for the case studied in ref. [2], namely, mixtures constituted by particles of the same mass density and equal total volumes of large and small particles. The data points represent the simulation results for $\alpha = 0.78$ when convection is suppressed. Points below (above) the curve correspond to RBNE (BNE).

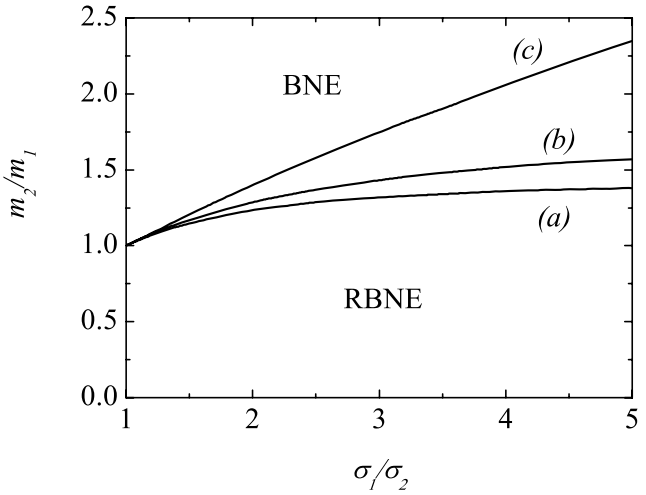

Fig. 3 – Phase diagram for BNE/RBNE in three dimensions for $\alpha_{ij} = 0.7$ and three values of composition: (a) $x_1 = 0$, (b) $x_1 = 0.3$, and (c) $x_1 = 0.7$. Points below (above) each curve correspond to RBNE (BNE).

small diameter ratios. However, since non-equipartition grows with increasing diameter ratio, the system shows a crossover to BNE at sufficiently large diameter ratios. This behavior agrees qualitatively well with the results reported in ref. [2] at large shaking amplitudes where thermal diffusion becomes the relevant segregation mechanism. At a quantitative level, we observe that the results are also consistent with the simulation results reported in [2] when periodic boundary conditions are used to suppress convection since they do not observe a change back to BNE for diameter ratios up to 3 (see red squares in fig. 11 of [2]). Although the parameter range explored in MD simulations is smaller than the one analyzed here, one is tempted to extrapolate the simulation data presented in ref. [2] to roughly predict the transition value of the diameter ratio at $\alpha = 0.78$ (which is the value of the coefficient of restitution considered in the simulations). Thus, if one extrapolates from the simulation data at the diameter ratios of 2 and 3, one sees that the transition from RBNE to BNE might be around $\sigma_1/\sigma_2 = 10$, which would quantitatively agree with the results reported in fig. 2. Figure 2 also shows that the BNE is completely destroyed in the quasielastic limit ($\alpha \simeq 1$).

Let us now investigate the influence of composition on segregation. Figure 3 shows a typical phase diagram in the three-dimensional case for $\alpha_{ij} \equiv \alpha = 0.7$ and three different values of the mole fraction x_1. The lines separate the regimes between BNE and RBNE. We observe that the composition of the mixture has significant effects in reducing the BNE as the concentration of larger particles increases. In addition, for a given value of composition, the transition from BNE to RBNE may occur following two paths: i) along the constant mass ratio m_1/m_2 with increasing size ratio σ_1/σ_2, and ii) along the constant size ratio with increasing mass ratio m_2/m_1. Another aim in this paper is to assess the influence of dissipation on the phase diagrams. Our results show that in general the role played by inelasticity is quite important since the regime of RBNE increases significantly with dissipation.

In summary, thermal diffusion (which is the relevant segregation mechanism in agitated granular mixtures at large shaking amplitudes) has been analyzed in the context of the inelastic Boltzmann equation by using a kinetic theory that is not restricted to small dissipation

and accounts for the non-equipartition of energy. The results reported here have been mainly motivated by recent experimental results [2] focussed on the analysis of three different segregation mechanisms: void filling, convection, and thermal diffusion. Concerning the latter effect, our model is able to explain some of the experimental and/or MD segregation results observed within the range of parameter space explored. In addition, the theoretical predictions for the temperature ratio of the mixture obtained from the driven condition (5) agree very well with computer simulations. A more quantitative comparison with MD simulations in the low-density regime is needed to show the relevance of the present theory. As said before, comparison with MD simulations in the tracer limit case ($x_1 \to 0$) [10] has shown the reliability of the inelastic Boltzmann equation to describe segregation. Given that the results derived here extends the description made in ref. [10] to arbitrary values of composition, one expects that such good agreement is also maintained for finite values of x_1. In this context, it is hoped that the present results stimulates the performance of such computer simulations in the dilute regime.

* * *

This research has been supported by the Ministerio de Educación y Ciencia (Spain) through grant No. FIS2004-01399, partially financed by FEDER funds.

REFERENCES

[1] See, for instance, Kudrolli A., *Rep. Prog. Phys.*, **67** (2004) 209.
[2] Schröter M., Ulrich S., Kreft J., Swift J. B. and Swinney H. L., to be published in *Phys. Rev. E*, and cond-mat/0601179 preprint (2006).
[3] Garzó V. and Dufty J. W., *Phys. Fluids*, **14** (2002) 1476; Garzó V., Montanero J. M. and Dufty J. W., cond-mat/0602198 preprint (2006); Garzó V. and Montanero J. M., cond-mat/0604552 preprint (2006).
[4] Dahl S. R., Hrenya C. M., Garzó V. and Dufty J. W., *Phys. Rev. E*, **66** (2002) 041301.
[5] Hong D. C., Quinn P. V. and Luding S., *Phys. Rev. Lett.*, **86** (2001) 3423.
[6] Jenkins J. T. and Yoon D., *Phys. Rev. Lett.*, **88** (2002) 194301.
[7] Breu A. P. J., Ensner H. M., Kruelle, C. A. and Rehberg I., *Phys. Rev. Lett.*, **90** (2003) 014302.
[8] Schautz T., Brito R., Kruelle C. A. and Rehberg I., *Phys. Rev. Lett.*, **95** (2005) 028001 and references therein.
[9] Trujillo L., Alam M. and Herrmann H. J., *Europhys. Lett.*, **64** (2003) 190.
[10] Brey J. J., Ruiz-Montero M. J. and Moreno F., *Phys. Rev. Lett.*, **95** (2005) 098001.
[11] Garzó V. and Dufty J. W., *Phys. Rev. E*, **60** (1999) 5706.