

Diffusion of intruders in a granular gas thermostatted by a bath of elastic hard spheres

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The Boltzmann kinetic equation is considered to compute the transport coefficients associated with the mass flux of intruders in a granular gas. Intruders and granular gas are immersed in a gas of elastic hard spheres (molecular gas). We assume that the granular particles are sufficiently rarefied so that the state of the molecular gas is not affected by the presence of the granular gas. Thus, the gas of elastic hard spheres can be considered as a thermostat (or bath) at a fixed temperature T_g . In the absence of spatial gradients, the system achieves a steady state where the temperature of the granular gas T differs from that of the intruders T_0 (energy non-equipartition). Approximate theoretical predictions for the temperature ratio T_0/T_g and the kurtosis c_0 associated with the intruders compare very well with Monte Carlo simulations for conditions of practical interest. For states close to the steady homogeneous state, the Boltzmann equation for the intruders is solved by means of the Chapman–Enskog method to first order in the spatial gradients. As expected, the diffusion transport coefficients are given in terms of the solutions of a set of coupled linear integral equations which are approximately solved by considering the first Sonine approximation. In dimensionless form, the transport coefficients are nonlinear functions of the mass and diameter ratios, the coefficients of restitution and the (reduced) bath temperature. Interestingly, previous results derived from a suspension model based on an effective fluid–solid interaction force are recovered when $m/m_g \rightarrow \infty$ and $m_0/m_g \rightarrow \infty$, where m , m_0 and m_g are the masses of the granular particles, intruders and molecular gas particles, respectively. Finally, as an application of our results, thermal diffusion segregation is exhaustively analysed.

Key words: granular media, gas/liquid flow

1. Introduction

One of the most relevant characteristics of granular systems is that they are constituted of macroscopic particles (or grains) that collide inelastically among themselves. Due to this fact, the kinetic energy of the system decreases over time. Thus, to observe sustained diffusive motion of the grains, an external energy input is usually introduced to compensate for the energy lost by collisions and reach a non-equilibrium steady state. Several mechanisms are used to inject energy to a system in real experiments, e.g. mechanical boundary shaking (Yang *et al.* 2002; Huan *et al.* 2004), bulk driving (as in air-fluidised beds (Schröter, Goldman & Swinney 2005; Abate & Durian 2006)) or magnetic forces (Sack *et al.* 2013; Harth *et al.* 2018). However, since in most experimental realisations the formation of large spatial gradients in the bulk region goes beyond the Navier–Stokes domain, it is quite difficult to provide a rigorous theoretical treatment of these sorts of situations. In computer simulations, the above obstacle can be circumvented by the introduction of external forces (or thermostats) (Evans & Morriss 1990) that heat the system and compensate for the energy dissipated by collisions. Unfortunately, it is not clear so far what the relation is between each specific type of thermostat and experiments.

A more realistic example of thermostatted granular systems consists of a set of solid particles immersed in an interstitial fluid of molecular particles. This provides a suitable starting point to mimic the behaviour of real suspensions. Needless to say, the understanding of the flow of solid particles in one or more fluid phases is in fact quite an intricate problem. Among the different types of multiphase flows, a simple but interesting set corresponds to the so-called particle-laden suspensions (Subramaniam 2020). In this sort of suspension, a set of small and dilute particles are immersed in a carrier fluid (such as water or air). When the dynamics of grains in gas–solid flows is essentially dominated by collisions, the extension of the conventional kinetic theory of gases (Chapman & Cowling 1970; Ferziger & Kaper 1972) to dissipative dynamics can be considered as a reliable tool to describe this sort of system. However, at a kinetic level, the description of flows involving two or more phases is really a complex problem since one should start from a set of kinetic equations for each one of the velocity distribution functions of the different phases. In addition, the different phases evolve over quite different spatial and temporal scales. Due to these difficulties, a coarse-grained approach is usually adopted and the influence of gas-phase effects on the dynamics of solid particles is incorporated in the starting kinetic equation in an effective way by means of a fluid–solid interaction force (Koch 1990; Gidaspow 1994; Jackson 2000). In some cases, a Stokes linear drag law for gas–solid interactions is only accounted for (Louge, Mastorakos & Jenkins 1991; Tsao & Koch 1995; Sangani *et al.* 1996; Wylie *et al.* 2009; Heussinger 2013; Wang *et al.* 2014; Chamorro, Vega Reyes & Garzó 2015; Saha & Alam 2017; Alam, Saha & Gupta 2019; Saha & Alam 2020; Chassagne, Bonamy & Chauchat 2023). Other models include an additional Langevin stochastic term (Garzó *et al.* 2012; Hayakawa, Takada & Garzó 2017; Gómez González & Garzó 2019; Gómez González, Khalil & Garzó 2020; Garzó 2023).

Although the effective suspension models based on the Langevin-like equation provide a reliable way of capturing the impact of gas-phase effects on the dynamic properties of grains, it could be convenient from a more fundamental point of view to begin with a model that accounts for the effect of the (real) collisions between solid and gas particles. In this context, inspired by a paper by Biben, Martin & Piasecki (2002), a (discrete) suspension model has been recently proposed (Gómez González & Garzó 2022b). As in the case of the most effective models reported in the granular literature (Louge *et al.* 1991; Tsao & Koch 1995; Sangani *et al.* 1996; Wylie *et al.* 2009; Garzó *et al.* 2012; Heussinger 2013; Wang *et al.* 2014; Chamorro *et al.* 2015; Hayakawa *et al.* 2017; Saha & Alam 2017;

Alam *et al.* 2019; Saha & Alam 2020; Gómez González & Garzó 2019; Gómez González *et al.* 2020), the model is based on the following assumptions. First, one assumes that the granular particles are sufficiently dilute so that the state of the interstitial gas is not affected by the presence of the grains. This means that the molecular surrounding gas can be treated as a bath (or thermostat) of elastic hard spheres at a constant temperature T_g . This assumption can be clearly justified in the case of particle-laden suspensions where the granular particles are sufficiently rarefied. Second, although the density of solid particles is very small, the grain–grain collisions (which are inelastic and characterised by the coefficient of restitution α) are accounted for in the kinetic equation for the velocity distribution function $f(\mathbf{r}, \mathbf{v}, t)$ of grains. Thus, it is quite obvious that this suspension model (granular particles immersed in a molecular gas of elastic hard spheres) can be seen as a binary mixture where one of the species (the grains) is present in tracer concentration. In the homogeneous state, a steady state is reached when the energy lost by grains (due to their inelastic collisions) is exactly compensated for by the energy gained by them due to their elastic collisions with gas particles (Biben *et al.* 2002; Santos 2003).

It is worth noting that the suspension model introduced by Gómez González & Garzó (2022*b*) has some features in common with the microscopic theory of transport for dilute molecular suspensions, as reported by Sung & Stell (1984*a,b*) years ago. In this theory, the dynamics of the solute–solvent collision is treated within the Enskog approximation, conveniently modified by the presence of the solvent sea. The solvent is treated as a continuum using appropriate generalised boundary conditions. These conditions allow the diffusion coefficient to properly account for dynamic memory (repeated collision events), which is neglected in the conventional Enskog theory. Additionally, the solute particles are sufficiently dilute so that the interaction between them may be neglected, yet concentrated enough to permit a statistical treatment. The theoretical expression for the self-diffusion coefficient is in excellent agreement with molecular dynamics simulations (Alder, Gass & Wainwright 1970). However, the microscopic theory of Sung & Stell (1984*a,b*) differs from the suspension model employed by Gómez González & Garzó (2022*b*). Firstly, the theory of Sung & Stell (1984*a,b*) is for elastic collisions, whereas the model of Gómez González & Garzó (2022*b*) considers the effect of the inelastic collisions between the solute (grains) particles on the distribution function. Secondly, the theory of Sung & Stell (1984*a,b*) considers finite values of the solid volume fraction of the solvent, whereas the suspension model introduced by Gómez González & Garzó (2022*b*) is restricted to the low-density regime. In this density regime, the inelastic Boltzmann kinetic equation applies, and it is justified to neglect the effect of dynamic correlations in repeated collisions on the transport coefficients. In this context, it is important to recall that for moderate densities the corresponding version of the inelastic Enskog equation (which goes beyond the Boltzmann description) can still be considered as a good approximation for obtaining the transport coefficients of dense granular fluids since the Enskog results (Garzó & Dufty 1999*a,b*; Garzó *et al.* 2007*a,b*) have been shown to compare quite well with molecular dynamics simulations (Lutsko, Brey & Dufty 2002; Dahl *et al.* 2002; Lois, Lemaître & Carlson 2007; Mitrano *et al.* 2011; Chialvo & Sundaresan 2013; Mitrano, Garzó & Hrenya 2014) and with experimental data (Yang *et al.* 2002; Huan *et al.* 2004) for moderately high densities and values of $\alpha \gtrsim 0.8$.

In contrast to coarse-grained approaches for granular suspensions, the model proposed by Gómez González & Garzó (2022*b*) introduces two new input parameters: the diameter σ/σ_g and mass m/m_g ratios. Here, σ_g and m_g are the diameter and mass of the particles of the surrounding molecular gas, respectively, while σ and m are the diameter and mass of the solid particles, respectively. For small spatial gradients, this suspension model has

been solved by means of the Chapman–Enskog method (Chapman & Cowling 1970) and the expressions of the Navier–Stokes transport coefficients of the granular suspension have been explicitly obtained in terms of the parameter space of the system (Gómez González & Garzó 2022b). An interesting result is that the Navier–Stokes expressions derived from this collisional model reduce to those previously derived from a coarse-grained approach (Gómez González & Garzó 2019) when the particles of the molecular gas are much lighter than the granular particles (Brownian limit, $m_g/m \rightarrow 0$). This agreement may justify the use of this sort of effective Langevin-like model for obtaining the dynamic properties of grains when $m \gg m_g$ (Pelargonio & Zaccone 2023).

While the study of transport properties in granular suspensions reported in Gómez González & Garzó (2022b) has been restricted to a monocomponent granular suspension (granular gas immersed in a molecular gas), extending this analysis to the more realistic case of a bidisperse granular suspension presents significant new conceptual and technical difficulties and is far from straightforward. Indeed, the evaluation of Navier–Stokes transport coefficients for multicomponent suspensions introduces significant new challenges. Not only does the number of relevant transport coefficients increase due to the complexity of particle interactions in mixtures, but also these coefficients are defined by a set of coupled integro-differential equations. Furthermore, new parameters emerge, such as the mass and size ratios, along with the coefficients of restitution for each pairwise collision, making the problem substantially more intricate than in the monocomponent case. Thus, to gain some insight into the general problem, we make in this paper a first step in the understanding of transport in multicomponent granular suspensions: we consider a granular binary mixture (immersed in a molecular gas) where the concentration of one of species (impurities or intruders) is much smaller than the other one (tracer limit). As mentioned before, in the tracer limit one can assume that (i) the state of the excess species (granular gas) is not perturbed by the presence of intruders and (ii) one can also neglect collisions among tracer particles themselves in their corresponding kinetic equation.

At a kinetic level, the tracer limit greatly simplifies the application of the Chapman–Enskog method (Chapman & Cowling 1970) to bidisperse granular suspensions since the transport properties of the excess species (the pressure tensor and the heat flux) are the same as those for the monocomponent granular suspension. These transport coefficients were already derived by Gómez González & Garzó (2022b). Consequently, the mass transport of impurities j_0 is the relevant flux of the problem. In accordance with the results of tracer diffusion in granular gases (Garzó 2019), one expects that the Navier–Stokes constitutive equation for the mass flux (that is, linear in the spatial gradients) can be written as

$$j_0^{(1)} = -\frac{m_0^2}{\rho} D_0 \nabla n_0 - \frac{mm_0}{\rho} D \nabla n - \frac{\rho}{T} D_T \nabla T - D_0^U \Delta U, \quad (1.1)$$

where $\rho = mn$ is the mass density of the granular gas, n_0 is the number density of the intruders, n is the number density of the particles of the granular gas, T is the granular temperature and $\Delta U = U - U_g$, U and U_g being the mean flow velocities of the granular and molecular gases, respectively. In addition, D_0 is the kinetic (tracer) diffusion coefficient, D is the mutual diffusion coefficient, D_T is the thermal diffusion coefficient and D_0^U is the velocity diffusion coefficient. While the first three diffusion coefficients are the coefficients of proportionality between the mass flux and hydrodynamic gradients, the coefficient D_0^U links the mass flux with the velocity difference ΔU . Although this latter contribution to the mass flux does not appear in dry granular mixtures, it is also present in the heat flux of a granular suspension composed of two different phases (a granular gas

immersed in a molecular gas) (Gómez González & Garzó 2022b). Here, as is shown later, by symmetry reasons the mass flux $j_0^{(1)}$ is also expected to be coupled to ΔU .

The determination of the diffusion transport coefficients D_0 , D , D_T and D_0^U is the main goal of the present paper. As usual for elastic (Chapman & Cowling 1970) and inelastic (Garzó 2019) collisions, these transport coefficients are given in terms of a set of coupled linear integral equations (see the supplementary material available at <https://doi.org/10.1017/jfm.2025.10806>). These integral equations are approximately solved by considering the leading terms in a Sonine polynomial expansion. However, as occurs in the case of a monocomponent granular suspension (Gómez González & Garzó 2022b), evaluating the diffusion coefficients for general unsteady conditions requires numerically solving a set of nonlinear differential equations. In the bidisperse case, these equations differ fundamentally from those of the monocomponent case due to the presence of two mechanically different species, resulting in the emergence of additional parameters. To simplify the analysis and obtain analytical results, we focus here on steady-state conditions. This enables us to get analytical results and express the diffusion transport coefficients in terms of the parameter space of the system.

The above set of diffusion transport coefficients has been recently determined in two different systems. Thus, in Gómez González *et al.* (2024) we considered a collisional model (the so-called Δ -model) to analyse the density flux of tracer particles in a confined, quasi-two-dimensional, moderately dense granular gas of inelastic hard spheres. More relevant to the present work, the diffusion coefficients of a binary granular suspension where one of the species (of mass m_0) is present in tracer concentration have been determined by solving the set of (inelastic) Enskog equations (Gómez González & Garzó 2023). In contrast to the suspension model considered here, a coarse-grained approach was adopted, whereby the influence of the interstitial fluid on grain motion was accounted for via effective forces (Langevin-like model). This simplification allowed us to derive explicit forms for the diffusion coefficients up to the second Sonine approximation. When m_0 is much greater than m_g , the results obtained in this study (which apply for arbitrary values of the mass ratio m_0/m_g) reduce to those derived in Gómez González & Garzó (2023) in the low-density regime and when only the first Sonine approximation is considered. In this sense, the present work subsumes previous studies (Gómez González & Garzó 2023) that are recovered in some limiting cases ($m_0/m_g \rightarrow \infty$ and $m/m_g \rightarrow \infty$).

Given that the explicit forms of the diffusion coefficients are at hand, as an interesting application of our results, we derive a segregation criterion for the intruders based on the knowledge of the so-called thermal diffusion factor (see e.g. Grew & Ibbs 1952; Goldhirsch & Ronis 1983a,b; Kincaid, Cohen & López de Haro 1987; Brey, Ruiz-Montero & Moreno 2005; Garzó 2006, 2008; Brito & Soto 2009; Gómez González & Garzó 2023; Gómez González *et al.* 2024). Segregation is induced here by both gravity and a temperature gradient. Three different situations are considered: one without gravity, another dominated by gravity and an intermediate case. Surprisingly, the segregation dynamics found here differs from that derived by using a Langevin-like approach in Gómez González & Garzó (2023). However, despite the plots appearing so different, we can explain those differences. They stem essentially from the way the molecular gas thermalises the grains in our (discrete) suspension model, which contrasts with the effective thermostat used in the coarse-grained approaches (Gómez González & Garzó 2023). Additionally, our model captures the full mass ratio dependence and therefore reveals how segregation varies as a function of the mass ratios m_0/m_g and m/m_g , offering a more general description beyond the Brownian limit ($m_0/m_g \rightarrow \infty$ and $m/m_g \rightarrow \infty$) considered in Gómez González & Garzó (2023). This is in fact one of the new added values of the present work.

The structure of the paper is as follows. Section 2 introduces the Boltzmann kinetic equation for granular particles immersed in a molecular gas and analyses the homogeneous steady state (HSS). In §3, some intruders are added to the granular gas and the corresponding Boltzmann–Lorentz kinetic equation is derived. We first consider the HSS for intruders and show how the non-equipartition of energy is affected by the mass ratio m_0/m_g . Section 4 presents the set of integral equations governing the diffusion transport coefficients, while §5 provides approximate expressions (based on the so-called first Sonine approximation) for these coefficients. These coefficients are explicitly determined in terms of the background temperature, volume fraction, restitution coefficients and the masses and diameters of the bidisperse system. Six appendices in the supplementary material provide technical details of the calculations and simulation techniques. The convergence to the results obtained by Gómez González & Garzó (2022a) from the Langevin-like model is also demonstrated. Section 7 examines thermal diffusion segregation. The paper concludes in §8 with a brief discussion of the results reported in this paper.

2. Granular gas in contact with a bath of elastic hard spheres. Boltzmann kinetic description

We consider a gas of inelastic hard disks ($d = 2$) or spheres ($d = 3$) of mass m , diameter σ and coefficient of normal restitution α . We assume that the spheres are perfectly smooth and therefore the collisions between particles are inelastic and characterised by a (positive) constant coefficient of normal restitution $\alpha \leq 1$. For elastic collisions $\alpha = 1$, while $\alpha < 1$ for inelastic collisions. The granular gas is immersed in a molecular gas consisting of hard disks or spheres of mass m_g and diameter σ_g . The collisions between the granular particles and the gas molecules are assumed to be elastic. As mentioned in §1, we also assume that the number density of the granular particles is much smaller than that of the molecular gas, so that the state of the latter is not significantly affected by the presence of grains. In this sense, the molecular gas can be treated as a thermostat or bath in equilibrium at temperature T_g . Thus, its velocity distribution function $f_g(\mathbf{V}_g)$ is

$$f_g(\mathbf{V}_g) = n_g \left(\frac{m_g}{2\pi T_g} \right)^{d/2} \exp \left(- \frac{m_g \mathbf{V}_g^2}{2T_g} \right), \quad (2.1)$$

where n_g is the number density of molecular gas and $\mathbf{V}_g = \mathbf{v} - \mathbf{U}_g$. In principle, the mean flow velocity of molecular gas $\mathbf{U}_g$ is different from the mean flow velocity $\mathbf{U}$ of solid particles (see its definition in (2.8)). In addition, for the sake of simplicity, we take the Boltzmann constant $k_B = 1$ throughout the paper.

In the low-density regime, the velocity distribution function $f(\mathbf{r}, \mathbf{v}, t)$ of granular particles verifies the Boltzmann kinetic equation. Moreover, although the granular gas is sufficiently rarefied and hence the properties of the molecular (interstitial) gas can be supposed to be constant, one has to take into account the collisions among grains themselves in the kinetic equation of $f(\mathbf{r}, \mathbf{v}, t)$. Thus, in the presence of the gravitational field $\mathbf{g}$, the distribution f verifies the Boltzmann equation:

$$\frac{\partial f}{\partial t} + \mathbf{v} \cdot \nabla f + \mathbf{g} \cdot \frac{\partial f}{\partial \mathbf{v}} = J[f, f] + J_g[f, f_g]. \quad (2.2)$$

Here, the Boltzmann collision operator $J[f, f]$ gives the rate of change of f due to inelastic collisions among granular particles. Its explicit form is (Brilliantov & Pöschel

2004; Garzó 2019)

$$J[\mathbf{v}_1|f, f] = \sigma^{d-1} \int d\mathbf{v}_2 \int d\hat{\boldsymbol{\sigma}} \Theta(\hat{\boldsymbol{\sigma}} \cdot \mathbf{g}_{12})(\hat{\boldsymbol{\sigma}} \cdot \mathbf{g}_{12}) \left[\alpha^{-2} f(\mathbf{v}_1'') f(\mathbf{v}_2'') - f(\mathbf{v}_1) f(\mathbf{v}_2) \right], \quad (2.3)$$

where $\mathbf{g}_{12} = \mathbf{v}_1 - \mathbf{v}_2$ is the relative velocity, $\hat{\boldsymbol{\sigma}}$ is a unit vector that joins the centres of the colliding particles and Θ is the Heaviside step function. In (2.3), the double primes denote pre-collisional velocities. The relation between them and their corresponding post-collisional velocities ($\mathbf{v}_1, \mathbf{v}_2$) is

$$\mathbf{v}_1'' = \mathbf{v}_1 - \frac{1+\alpha}{2\alpha} (\hat{\boldsymbol{\sigma}} \cdot \mathbf{g}_{12}) \hat{\boldsymbol{\sigma}}, \quad \mathbf{v}_2'' = \mathbf{v}_2 + \frac{1+\alpha}{2\alpha} (\hat{\boldsymbol{\sigma}} \cdot \mathbf{g}_{12}) \hat{\boldsymbol{\sigma}}. \quad (2.4)$$

In (2.2), the Boltzmann–Lorentz operator $J_g[f, f_g]$ accounts for the rate of change of f due to elastic collisions between particles of the granular and molecular gas. Its form is (R  sibois & de Leener 1977)

$$J_g[\mathbf{v}_1|f, f_g] = \bar{\sigma}^{d-1} \int d\mathbf{v}_2 \int d\hat{\boldsymbol{\sigma}} \Theta(\hat{\boldsymbol{\sigma}} \cdot \mathbf{g}_{12})(\hat{\boldsymbol{\sigma}} \cdot \mathbf{g}_{12}) \left[f(\mathbf{v}_1'') f_g(\mathbf{v}_2'') - f(\mathbf{v}_1) f_g(\mathbf{v}_2) \right], \quad (2.5)$$

where $\bar{\sigma} = (\sigma + \sigma_g)/2$ and the relationship between ($\mathbf{v}_1'', \mathbf{v}_2''$) and ($\mathbf{v}_1, \mathbf{v}_2$) is

$$\mathbf{v}_1'' = \mathbf{v}_1 - 2\mu_g (\hat{\boldsymbol{\sigma}} \cdot \mathbf{g}_{12}) \hat{\boldsymbol{\sigma}}, \quad \mathbf{v}_2'' = \mathbf{v}_2 + 2\mu (\hat{\boldsymbol{\sigma}} \cdot \mathbf{g}_{12}) \hat{\boldsymbol{\sigma}}. \quad (2.6)$$

Here,

$$\mu_g = \frac{m_g}{m + m_g}, \quad \mu = \frac{m}{m + m_g}. \quad (2.7)$$

As is customary, the effect of gravity on the properties of molecular gases under ordinary conditions is neglected in the present analysis. This approximation is justified by the fact that the influence of gravity on a molecule between successive collisions is negligible, i.e. $\ell \ll h$, where ℓ denotes the mean free path for hard spheres and $h = v_{\text{th}}^2/g$ represents the characteristic length scale over which gravitational effects become significant (v_{th} being the thermal velocity). For instance, under terrestrial conditions at room temperature, this ratio is of the order of $\ell/h \sim 10^{-11}$ (Tij, Garz   & Santos 1999), thereby validating the omission of gravitational effects in the description of molecular gas behaviour.

The number density n , mean flow velocity $\mathbf{U}$ and granular temperature T of the granular gas are defined as the first few velocity moments of f :

$$\{n, n\mathbf{U}, dnT\} = \int d\mathbf{v} \left\{ 1, \mathbf{v}, mV^2 \right\} f(\mathbf{v}), \quad (2.8)$$

where $\mathbf{V} = \mathbf{v} - \mathbf{U}$ is the peculiar velocity. As said before, the difference $\Delta\mathbf{U} = \mathbf{U} - \mathbf{U}_g$ is in general different from zero (G  mez Gonz  lez & Garz   2022b). In fact, as we show later, $\Delta\mathbf{U}$ induces a non-vanishing contribution to the mass flux of intruders.

The macroscopic balance equations for the granular gas are obtained by multiplying (2.2) by $\{1, \mathbf{v}, mV^2\}$ and integrating over velocity. The result is (G  mez Gonz  lez & Garz   2022b)

$$D_t n + n \nabla \cdot \mathbf{U} = 0, \quad (2.9)$$

$$\rho D_t \mathbf{U} = -\nabla \cdot \mathbf{P} + \rho \mathbf{g} + \mathcal{F}[f], \quad (2.10)$$

$$D_t T + \frac{2}{dn} (\nabla \cdot \mathbf{q} + \mathbf{P} : \nabla \mathbf{U}) = -T\zeta - T\zeta_g. \quad (2.11)$$

In (2.9)–(2.11), $D_t = \partial_t + \mathbf{U} \cdot \nabla$ is the material derivative and the pressure tensor $\mathbf{P}$ and the heat flux vector $\mathbf{q}$ are given, respectively, as

$$\mathbf{P} = \int d\mathbf{v} m \mathbf{V} \mathbf{V} f(\mathbf{v}), \quad \mathbf{q} = \int d\mathbf{v} \frac{m}{2} V^2 \mathbf{V} f(\mathbf{v}). \quad (2.12)$$

The production of momentum term $\mathcal{F}[f]$ appearing in (2.10) is defined as

$$\mathcal{F}[f] = \int d\mathbf{v} m \mathbf{V} J_g[f, f_g]. \quad (2.13)$$

This term is in general different from zero since the Boltzmann–Lorentz collision operator $J_g[f, f_g]$ does not conserve momentum. The form of $\mathcal{F}[f]$ can be made more explicit when one takes into account the property (Brilliantov & Pöschel 2004; Garzó 2019)

$$\begin{aligned} \int d\mathbf{v}_1 \Psi(\mathbf{v}_1) J_g[\mathbf{v}_2|f, f_g] &= \bar{\sigma}^{d-1} \int d\mathbf{v}_1 \int d\mathbf{v}_2 \int d\hat{\boldsymbol{\sigma}} \Theta(\hat{\boldsymbol{\sigma}} \cdot \mathbf{g}_{12}) (\hat{\boldsymbol{\sigma}} \cdot \mathbf{g}_{12}) f(\mathbf{v}_1) f_g(\mathbf{v}_2) \\ &\times [\Psi(\mathbf{v}'_1) - \Psi(\mathbf{v}_1)], \end{aligned} \quad (2.14)$$

where $\mathbf{v}'_1 = \mathbf{v}_1 - 2\mu_g(\hat{\boldsymbol{\sigma}} \cdot \mathbf{g}_{12})\hat{\boldsymbol{\sigma}}$. Using (2.14), $\mathcal{F}[f]$ is

$$\mathcal{F}[f] = -\frac{2\pi^{(d-1)/2}}{\Gamma\left(\frac{d+3}{2}\right)} \frac{mm_g}{m+m_g} \bar{\sigma}^{d-1} \int d\mathbf{v}_1 \int d\mathbf{v}_2 g_{12} \mathbf{g}_{12} f(\mathbf{v}_1) f_g(\mathbf{v}_2). \quad (2.15)$$

Finally, the partial production rates ζ and ζ_g appearing in the balance equation (2.11) are given, respectively, as

$$\zeta = -\frac{m}{dnT} \int d\mathbf{v} V^2 J[\mathbf{v}|f, f], \quad \zeta_g = -\frac{m}{dnT} \int d\mathbf{v} V^2 J_g[\mathbf{v}|f, f_g]. \quad (2.16)$$

While the cooling rate ζ provides the rate of change of kinetic energy of grains due to their inelastic collisions, the term ζ_g gives the transfer of kinetic energy in the collisions between the particles of the molecular and granular gases. The quantity $\zeta = 0$ for elastic collisions ($\alpha = 1$) while $\zeta_g = 0$ when the particles of the molecular and granular gases are mechanically equivalent.

It is interesting at this point to note the meaning of the granular temperature T . To understand this, it is important to remember that our study is limited to the so-called rapid flow regime, namely a situation where grains are subjected to a strong external excitation (e.g. vibrating or shearing walls or air-fluidised beds). In this regime, the external energy supplied to the granular gas can compensate for the energy loss due to collisions and the effects of gravity. Since in this regime the motion of the grains is quite similar to the chaotic motion of atoms or molecules in an ordinary gas, as discussed in previous works (Gómez González & Garzó 2022b), it is tempting to establish a relationship between the statistical motion of the grains and some kind of temperature. In this context, as usual in conventional kinetic theory (Chapman & Cowling 1970), the granular temperature T can be interpreted as a measure of the fluctuations of the velocities of grains with respect to its mean value $\mathbf{U}$. Since granular gases are athermal systems (i.e. their thermal fluctuations have a negligible effect on the dynamics of grains), the granular temperature T has no thermodynamic interpretation in contrast to the temperature T_g of the molecular gas (see, for example, the review paper of Goldhirsch (2008) for a discussion of this issue). In any case, within the context of statistical thermodynamics, the thermodynamic temperature T_g can be also understood as a statistical quantity measuring the deviations of molecular particle velocity $\mathbf{v}$ from its mean value $\mathbf{U}_g$.

Before closing this section, it is worth analysing the limiting case $m \gg m_g$. It is in fact a quite realistic case for granular suspensions (Subramaniam 2020) where the particles of the interstitial molecular gas are much lighter than the particles of the granular gas. In the limit $m/m_g \rightarrow \infty$, a Kramers–Moyal expansion (R  sibois & de Leener 1977) allows us to approximate the Boltzmann–Lorentz operator $J_g[f, f_g]$ to the Fokker–Planck operator $J_g^{FP}[f, f_g]$:

$$J_g[f, f_g] \rightarrow J_g^{FP}[f, f_g] = \gamma \frac{\partial}{\partial \mathbf{v}} \cdot \left(\mathbf{v} + \frac{T_g}{m} \frac{\partial}{\partial \mathbf{v}} \right) f(\mathbf{v}), \quad (2.17)$$

where the friction coefficient γ is

$$\gamma = \frac{4\pi^{(d-1)/2}}{\Gamma(\frac{d}{2})} \left(\frac{m_g}{m} \right)^{1/2} \left(\frac{2T_g}{m} \right)^{1/2} n_g \bar{\sigma}^{d-1}. \quad (2.18)$$

Upon deriving (2.17), it has been assumed that $\mathbf{U}_g = \mathbf{0}$ and that the distribution function f of the granular gas is a Maxwellian distribution.

Most of the theoretical works for suspension models reported in the granular literature are essentially based on the use of the Fokker–Planck operator (2.17) to account for in an effective way (coarse-grained approach) the influence of the surrounding fluid on the dynamics of grains (Koch & Hill 2001; Garz   *et al.* 2012; Chassagne *et al.* 2023; Garz   2023). These sorts of models have been considered to obtain the Navier–Stokes–Fourier transport coefficients of the suspension (G  mez Gonz  lez & Garz   2019).

3. Intruders in granular suspensions

We assume now that a few intruders (or tracers) of mass m_0 and diameter σ_0 are added to the granular gas. In this situation, intruders and particles of the granular gas are surrounded by the molecular gas (bath of elastic hard spheres). The system can be seen as a ternary mixture where one of the components (intruders) is present in tracer concentration. Apart from the restitution coefficient α for inelastic grain–grain collisions, the coefficient of normal restitution $\alpha_0 \leq 1$ characterises the inelastic collisions between the intruders and the particles of the granular gas. As in the case of the granular gas, collisions between intruders and particles of the surrounding molecular gas are elastic.

Since the concentration of intruders is much smaller than that of the granular gas (tracer limit), their presence does not affect the state of the granular gas. Under these conditions and in the presence of the gravitational field, the velocity distribution function $f_0(\mathbf{r}, \mathbf{v}, t)$ of the intruders obeys the kinetic equation:

$$\frac{\partial f_0}{\partial t} + \mathbf{v} \cdot \nabla f_0 + \mathbf{g} \cdot \frac{\partial f_0}{\partial \mathbf{v}} = J_0[f_0, f] + J_{0g}[f_0, f_g], \quad (3.1)$$

where the (inelastic) version of the Boltzmann–Lorentz collision operator $J_0[f_0, f]$ gives the rate of change of f_0 due to the inelastic collisions between the intruders and particles of the granular gas. It is given by (Garz   2019)

$$J_0[\mathbf{v}_1|f_0, f] = \sigma'^{(d-1)} \int d\mathbf{v}_2 \int d\hat{\boldsymbol{\sigma}} \Theta(\hat{\boldsymbol{\sigma}} \cdot \mathbf{g}_{12})(\hat{\boldsymbol{\sigma}} \cdot \mathbf{g}_{12}) \left[\alpha_0^{-2} f_0(\mathbf{v}_1'') f(\mathbf{v}_2'') - f_0(\mathbf{v}_1) f(\mathbf{v}_2) \right]. \quad (3.2)$$

As in (2.3), $\mathbf{g}_{12} = \mathbf{v}_1 - \mathbf{v}_2$ is the relative velocity, $\hat{\boldsymbol{\sigma}}$ is a unit vector and Θ is the Heaviside step function. In addition, $\sigma' = (\sigma + \sigma_0)/2$,

$$\mathbf{v}_1'' = \mathbf{v}_1 - \frac{1 + \alpha_0}{\alpha_0} \mu'(\hat{\boldsymbol{\sigma}} \cdot \mathbf{g}_{12}) \hat{\boldsymbol{\sigma}}, \quad \mathbf{v}_2'' = \mathbf{v}_2 + \frac{1 + \alpha_0}{2\alpha_0} \mu'_0(\hat{\boldsymbol{\sigma}} \cdot \mathbf{g}_{12}) \hat{\boldsymbol{\sigma}} \quad (3.3)$$

and

$$\mu' = \frac{m}{m + m_0}, \quad \mu'_0 = \frac{m_0}{m + m_0}. \quad (3.4)$$

In (3.1), the collision operator $J_{0g}[f_0, f_g]$ provides the rate of change of f_0 due to elastic collisions between intruders and particles of the molecular gas. Similarly to the operator $J_0[f, f_g]$, it is given by

$$J_{0g}[\mathbf{v}_1 | f_0, f_g] = \bar{\sigma}_0^{d-1} \int d\mathbf{v}_2 \int d\hat{\boldsymbol{\sigma}} \Theta(\hat{\boldsymbol{\sigma}} \cdot \mathbf{g}_{12})(\hat{\boldsymbol{\sigma}} \cdot \mathbf{g}_{12}) [f_0(\mathbf{v}_1'') f_g(\mathbf{v}_2'') - f_0(\mathbf{v}_1) f_g(\mathbf{v}_2)]. \quad (3.5)$$

Here, $\bar{\sigma}_0 = (\sigma_0 + \sigma_g)/2$,

$$\mathbf{v}_1'' = \mathbf{v}_1 - 2\mu_{g0}(\hat{\boldsymbol{\sigma}} \cdot \mathbf{g}_{12})\hat{\boldsymbol{\sigma}}, \quad \mathbf{v}_2'' = \mathbf{v}_2 + 2\mu_{0g}(\hat{\boldsymbol{\sigma}} \cdot \mathbf{g}_{12})\hat{\boldsymbol{\sigma}} \quad (3.6)$$

and

$$\mu_{g0} = \frac{m_g}{m_0 + m_g}, \quad \mu_{0g} = \frac{m_0}{m_0 + m_g}. \quad (3.7)$$

Although the granular temperature T is the relevant one at a hydrodynamic level, an interesting quantity at a kinetic level is the local temperature of the intruders T_0 . This quantity measures the mean kinetic energy of the intruders. It is defined as

$$T_0(\mathbf{r}, t) = \frac{m_0}{dn_0(\mathbf{r}, t)} \int d\mathbf{v} V^2 f_0(\mathbf{r}, \mathbf{v}, t). \quad (3.8)$$

As confirmed by kinetic theory calculations (Garzó & Dufty 1999b), computer simulations (Garzó 2019) and experiments (Feitosa & Menon 2002; Wildman & Parker 2002; Puzyrev *et al.* 2024), the global temperature T and the temperature of impurities T_0 are in general different.

Intruders may freely exchange momentum and energy with the particles of the granular and molecular gas. Thus, only the number density of intruders

$$n_0(\mathbf{r}, t) = \int d\mathbf{v} f_0(\mathbf{r}, \mathbf{v}, t) \quad (3.9)$$

is conserved. This yields the balance equation

$$\frac{\partial \rho_0}{\partial t} + \nabla \cdot \mathbf{j}_0 = 0, \quad (3.10)$$

where $\rho_0 = m_0 n_0$ is the mass density of intruders and

$$\mathbf{j}_0(\mathbf{r}, t) = \int d\mathbf{v} m_0 \mathbf{v} f_0(\mathbf{r}, \mathbf{v}, t) \quad (3.11)$$

is the mass flux of intruders.

As in the case of the Boltzmann–Lorentz operator $J_g[f, f_g]$, in the limiting case $m_0 \gg m_g$ the operator $J_{0g}[f_0, f_g]$ reduces to the Fokker–Planck operator:

$$J_{0g}[f_0, f_g] \rightarrow J_{0g}^{FP}[f_0, f_g] = \gamma_0 \frac{\partial}{\partial \mathbf{v}} \cdot \left(\mathbf{v} + \frac{T_g}{m_0} \frac{\partial}{\partial \mathbf{v}} \right) f_0(\mathbf{v}), \quad (3.12)$$

where the friction coefficient γ_0 is

$$\gamma_0 = \frac{4\pi^{(d-1)/2}}{d\Gamma\left(\frac{d}{2}\right)} \left(\frac{m_g}{m_0}\right)^{1/2} \left(\frac{2T_g}{m_0}\right)^{1/2} n_g \bar{\sigma}_0^{d-1}. \quad (3.13)$$

Note that the expression (3.13) for γ_0 differs from the macroscopic Stokes law describing the Brownian motion of a massive particle in an equilibrium host fluid (Bocquet, Hansen & Piaseck 1994a,b; Gómez González & Garzó 2022a). This means that the results derived in this paper in the Brownian limit do not exactly reduce to those obtained by using a coarse-grained approach (Gómez González & Garzó 2022a).

4. Homogeneous steady state

Before considering inhomogeneous situations, it is pertinent to analyse the HSS for the system (intruders and granular gas immersed in a molecular gas) in the absence of the gravitational field ($\mathbf{g} = \mathbf{0}$). The study of this state for the intruders is crucial since the HSS plays the role of the reference state in the Chapman–Enskog method (Chapman & Cowling 1970).

In the HSS, the densities n and n_0 and the granular temperature T are spatially uniform. Moreover, without loss of generality, the mean flow velocities vanish ($\mathbf{U} = \mathbf{U}_g = \mathbf{0}$) with an appropriate choice of the frame of reference. Under these conditions, (2.2) for the granular gas reads

$$\partial_t f = J[f, f_0] + J_g[f, f_g], \quad (4.1)$$

while (3.1) for the intruders becomes

$$\partial_t f_0 = J_0[f_0, f] + J_{0g}[f_0, f_g]. \quad (4.2)$$

4.1. Homogeneous steady state for the granular gas

Given that the HSS for the granular gas was already analysed by Gómez González & Garzó (2022b), only a few results are provided here. Since in the HSS the distribution $f(\mathbf{v})$ is isotropic in $\mathbf{v}$, then $\mathcal{F}[f] = \mathbf{0}$. On the other hand, the kinetic energy is not conserved by collisions and so $\zeta \neq 0$ and $\zeta_g \neq 0$. The most interesting quantity in the HSS for the granular gas is the temperature ratio $\chi = T/T_g$, which is in general different from 1. The only non-trivial balance equation in the homogeneous state for the granular gas is that of the temperature (2.11):

$$\frac{\partial T}{\partial t} = -T (\zeta + \zeta_g). \quad (4.3)$$

After a transient period, it is expected that the granular gas achieves a steady state. Thus, according to (4.3), the steady-state condition is $\zeta + \zeta_g = 0$. As discussed by Gómez González & Garzó (2022b), as the molecular gas acts as a thermostat in the steady state, then the mean kinetic energy of the granular particles is smaller than that of the molecular gas ($T < T_g$). This necessarily requires that $\zeta_g < 0$ so that, in the steady state, the production rates ζ and $|\zeta_g|$ exactly compensate each other and one achieves the condition $\zeta + \zeta_g = 0$.

However, to determine ζ and ζ_g one needs to know the velocity distribution function $f(\mathbf{v})$ for the granular gas. For inelastic collisions ($\alpha < 1$) this distribution is not exactly known to date. On the other hand, the results obtained for the fourth cumulant or kurtosis c of the distribution f in Gómez González & Garzó (2022b) clearly show (see figure 3 of Gómez González & Garzó (2022b)) that the magnitude of c is in general very small for not quite strong inelasticity (e.g. $\alpha \gtrsim 0.5$). Thus, to estimate the production rates ζ and ζ_g one can replace the true distribution $f(\mathbf{v})$ by the Maxwellian distribution:

$$f_M(\mathbf{v}) = n \left(\frac{m}{2\pi T} \right)^{d/2} \exp \left(-\frac{m\mathbf{v}^2}{2T} \right). \quad (4.4)$$

In the Maxwellian approximation, the dimensionless production rates $\zeta^* = \zeta/\nu$ and $\zeta_g^* = \zeta_g/\nu$ are given by (Gómez González & Garzó 2022b)

$$\zeta^* = \frac{\sqrt{2}\pi^{(d-1)/2}}{d\Gamma\left(\frac{d}{2}\right)}(1-\alpha^2), \quad \zeta_g^* = 2x(1-x^2)\left(\frac{\mu T}{T_g}\right)^{1/2}\gamma^*, \quad (4.5)$$

where $\nu = n\sigma^{d-1}\sqrt{2T/m}$ is an effective collision frequency,

$$x = \left(\mu_g + \mu\frac{T_g}{T}\right)^{1/2} \quad (4.6)$$

and

$$\gamma^* = \epsilon\chi^{-1/2}, \quad \epsilon = \frac{\ell\gamma}{\sqrt{2T_g/m}} = \frac{\sqrt{2}\pi^{d/2}}{2^d d\Gamma\left(\frac{d}{2}\right)}\frac{1}{\phi\sqrt{T_g^*}}. \quad (4.7)$$

Here, $\ell = 1/n\sigma^{d-1}$ is proportional to the mean free path of hard spheres,

$$\phi = \frac{\pi^{d/2}}{2^{d-1}d\Gamma\left(\frac{d}{2}\right)}n\sigma^d \quad (4.8)$$

is the solid volume fraction and

$$T_g^* = \frac{T_g}{m\sigma^2\gamma^2}. \quad (4.9)$$

In the Maxwellian approximation (i.e. when one replaces f by f_M), the steady temperature ratio T/T_g can be obtained by inserting the expressions (4.5) of ζ^* and ζ_g^* , respectively, into the (exact) steady-state condition $\zeta^* + \zeta_g^* = 0$. This yields a cubic equation for the quantity x whose physical solution is given by (4.14) of the supplementary material of Gómez González & Garzó (2022b). In terms of x , the final expression of the temperature ratio T/T_g is given by (4.15) of that supplementary material. In spite of considering the Maxwellian approximation for the distribution f , an excellent agreement between theory and simulations for the temperature ratio is observed over the whole range of values of α studied (see figure 1 of Gómez González & Garzó (2022b)).

4.2. Homogeneous steady state for the intruders

We analyse now the HSS for the intruders. The balance equation for the intruders' temperature T_0 can be easily obtained from (4.2) as $\partial_t \ln T_0 = -(\zeta_0 + \zeta_{0g})$, where

$$\zeta_0 = -\frac{m_0}{dn_0T_0} \int d\mathbf{v} v^2 J_0[\mathbf{v}|f_0, f], \quad \zeta_{0g} = -\frac{m_0}{dn_0T_0} \int d\mathbf{v} v^2 J_{0g}[\mathbf{v}|f_0, f_g]. \quad (4.10)$$

In the HSS, $\partial_t T_0 = 0$ and the condition for obtaining T_0 is

$$\zeta_0 + \zeta_{0g} = 0. \quad (4.11)$$

As in the case of the granular gas, the exact form of the distribution function $f_0(\mathbf{v})$ for inelastic collisions is not known to date. The departure of $f_0(\mathbf{v})$ from its Maxwellian form

$$f_{0,M}(\mathbf{v}) = n_0 \left(\frac{m_0}{2\pi T_0}\right)^{d/2} \exp\left(-\frac{m_0 v^2}{2T_0}\right) \quad (4.12)$$

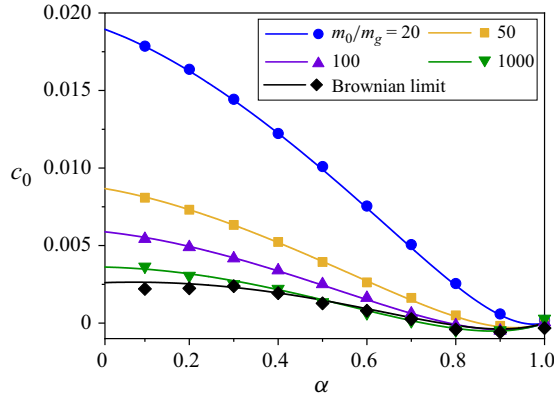


Figure 1. Plot of the kurtosis c_0 associated with the distribution function of the intruders as a function of the coefficient of normal restitution α for $d = 3$, $\phi = 0.0052$, $T_g^* = 1000$ and four different values of the mass ratio m_0/m_g (from top to bottom, $m_0/m_g = 20$, 50, 100 and 1000). Moreover, in all the curves $m_0/m = 10$, $\sigma_0/\sigma = 5$ and $\sigma_0/\sigma_g = (m_0/m_g)^{1/3}$. The solid lines are the theoretical results while the symbols are the DSMC simulation results. The dashed line is the result obtained from the Fokker–Planck approach (3.12) to the operator $J_{0g}[f_0, f_g]$. Diamonds refer to DSMC simulations implemented using the time-driven approach (Gómez González & Garzó 2022b).

can be measured by the kurtosis c_0 . It is defined as (Garzó 2019)

$$c_0 = \frac{1}{d(d+2)} \frac{m_0^2}{n_0 T_0^2} \int d\mathbf{v} v^4 f_0(\mathbf{v}) - 1. \quad (4.13)$$

Some technical details on the determination of c_0 are given in the supplementary material. Given that the expression of c_0 is very large and not very illuminating, its final form is not displayed here. In terms of dimensionless quantities, the parameter space of a d -dimensional system is given by the set

$$\xi \equiv \left\{ \frac{\sigma}{\sigma_g}, \frac{\sigma_0}{\sigma_g}, \frac{m}{m_g}, \frac{m_0}{m_g}, \alpha, \alpha_0, \phi, T_g^* \right\}. \quad (4.14)$$

In contrast to the monocomponent case (Gómez González & Garzó 2022b), note that the diameter ratios σ/σ_g and σ_0/σ_g appear also as input parameters of the system.

Figure 1 shows c_0 versus the (common) coefficient of restitution $\alpha = \alpha_0$ for $d = 3$, $\phi = 0.0052$ and $T_g^* = 1000$. Four different values of the mass ratio m_0/m_g (20, 50, 100 and 1000) are considered keeping the ratio $m_0/m = 10$. In addition, $\sigma_0/\sigma = 5$ and we have assumed that the intruders and molecular gas particles have the same mass density (i.e. $\sigma_0/\sigma = (m_0/m_g)^{1/3}$). As occurs for the kurtosis c of the granular gas (see figure 3 of Gómez González & Garzó (2022b)), it is quite apparent from figure 1 that the magnitude of c_0 is in general quite small. We observe that the agreement between theory and direct simulation Monte Carlo (DSMC) simulations is excellent even for quite extreme values of inelasticity.

The fact that c_0 is small allows us to guarantee that a good estimate of the production rates ζ_0 and ζ_{0g} can be obtained by replacing $f_0(\mathbf{v})$ by the Maxwellian distribution $f_{0,M}(\mathbf{v})$ in (4.10). In this approximation, the dimensionless quantities $\zeta_0^* = \zeta_0/\nu$ and $\zeta_{0g}^* = \zeta_{0g}/\nu$ can be written as

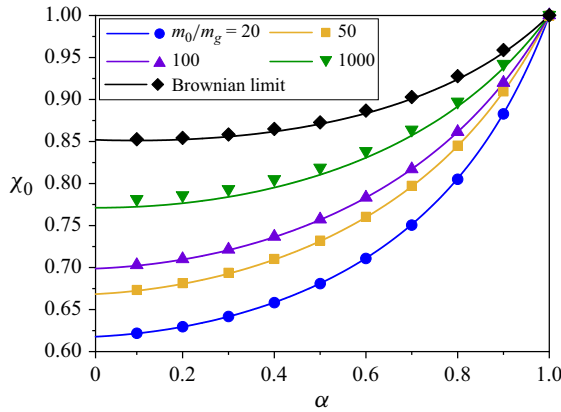


Figure 2. Temperature ratio $\chi_0 \equiv T_0/T_g$ versus the (common) coefficient of normal restitution $\alpha_0 = \alpha$ for $d = 3$, $\phi = 0.0052$, $T_g^* = 1000$ and four different values of the mass ratio m_0/m_g (from top to bottom, $m_0/m_g = 20, 50, 100$ and 1000). Moreover, in all the curves $m_0/m = 10$, $\sigma_0/\sigma = 5$ and $\sigma_0/\sigma_g = (m_0/m_g)^{1/3}$. The solid lines are the theoretical results while the symbols are the DSMC simulation results. The dashed line is the result obtained by using the Fokker–Planck approach (3.12) to the operator $J_{0g}[f_0, f_g]$. Diamonds refer to DSMC simulations implemented using the time-driven approach (Gómez González & Garzó 2022b).

$$\zeta_0^* = \frac{4\pi^{(d-1)/2}}{d\Gamma\left(\frac{d}{2}\right)} \mu' \left(\frac{\sigma'}{\sigma}\right)^{d-1} \left(1 + \frac{mT_0}{m_0T}\right)^{1/2} (1 + \alpha_0) \left[1 - \frac{\mu'}{2}(1 + \alpha_0) \left(1 + \frac{m_0T}{mT_0}\right)\right], \quad (4.15)$$

$$\zeta_{0g}^* = 2x_0(1 - x_0^2) \left(\mu_{0g} \frac{T_0}{T_g}\right)^{1/2} \gamma_0^*. \quad (4.16)$$

In (4.15) and (4.16), we have introduced the quantities

$$x_0 = \left(\mu_{g0} + \mu_{0g} \frac{T_g}{T_0}\right)^{1/2}, \quad (4.17)$$

$$\gamma_0^* = \epsilon_0 \chi^{-1/2}, \quad \epsilon_0 = \left(\frac{\bar{\sigma}_0}{\bar{\sigma}}\right)^{d-1} \frac{m}{m_0} \epsilon. \quad (4.18)$$

The temperature ratio $\chi_0 \equiv T_0/T_g$ can be finally determined by substituting (4.15) and (4.16) into the condition (4.11). It yields the nonlinear algebraic equation

$$2x_0(x_0^2 - 1) (\mu_{0g} \chi_0)^{1/2} \gamma_0^* = \frac{4\pi^{(d-1)/2}}{d\Gamma\left(\frac{d}{2}\right)} \mu' \left(\frac{\sigma'}{\sigma}\right)^{d-1} \left(1 + \frac{m\chi_0}{m_0\chi}\right)^{1/2} (1 + \alpha_0) \times \left[1 - \frac{\mu}{2}(1 + \alpha_0) \left(1 + \frac{m_0\chi}{m\chi_0}\right)\right], \quad (4.19)$$

where we recall that $\chi = T/T_g$ is given by (3.7) of the supplementary material of Gómez González & Garzó (2022b). The numerical solution to (4.17) provides the dependence of T_0/T_g on the parameter space ξ .

For the sake of illustration, figure 2 shows $\chi_0 \equiv T_0/T_g$ as a function of the (common) coefficient of restitution $\alpha_0 = \alpha$ for the same systems as in figure 1. As occurs for the ratio T/T_g , due to the way of scaling the relevant quantities of the system, the deviation of χ_0 from unity increases with decreasing the mass ratio m_0/m_g . The agreement between the (approximate) theoretical results and simulations is again excellent; it clearly justifies the

use of the Maxwellian distribution (4.12) to achieve accurate estimates of the cooling rates ζ_0 and ζ_{0g} .

A point to consider here is that convergence to the results obtained using the Fokker–Planck model is achieved not only in the limit $m/m_g \rightarrow \infty$ and $m_0/m_g \rightarrow \infty$ but it is also necessary that $\sigma/\sigma_g \rightarrow \infty$ and $\sigma_0/\sigma_g \rightarrow \infty$. For convenience, we assume in the rest of the work that $m/m_g \rightarrow \infty$ and $m_0/m_g \rightarrow \infty$ also imply $\sigma/\sigma_g \rightarrow \infty$ and $\sigma_0/\sigma_g \rightarrow \infty$, and thus intruders and molecular gas particles have the same particle mass density (i.e. $m_0/\sigma_0^d = m_g/\sigma_g^d$).

5. Chapman–Enskog method. Diffusion transport coefficients

We assume that we perturb the homogeneous state by small spatial gradients. These perturbations induce non-vanishing contributions to the mass, momentum and heat fluxes. The determination of these fluxes allows us to identify the corresponding Navier–Stokes–Fourier transport coefficients of the granular suspension. As said in § 1, since in the tracer limit the pressure tensor and the heat flux vector of the binary mixture (intruders plus granular gas) are the same as that for the excess species (granular gas), the mass transport of intruders $\mathbf{j}_0$ is the relevant flux of the problem. The Navier–Stokes–Fourier transport coefficients of the granular gas were already determined by Gómez González & Garzó (2022b).

To get the mass flux $\mathbf{j}_0$, the Boltzmann kinetic equation (3.1) is solved up to first order in spatial gradients by means of the Chapman–Enskog expansion Chapman & Cowling (1970) conveniently adapted to dissipative dynamics. As widely discussed in many textbooks (Chapman & Cowling 1970; Ferziger & Kaper 1972), there are two different stages in the relaxation of a molecular gas towards equilibrium. For times of the order of the mean free time, one can identify a first stage (kinetic stage) where the main effect of collisions on the distribution function is to relax it towards the so-called local equilibrium state. Then, a hydrodynamic (slow) stage is achieved where the gas has completely forgotten its initial preparation. In this stage, the microscopic state of the gas is completely specified by the knowledge of the hydrodynamic fields (in the case of a binary mixture by n_0 , n , $\mathbf{U}$ and T). These two stages are also expected in the case of granular gases except that in the kinetic stage the distribution function will generally relax towards a time-dependent non-equilibrium distribution (the homogeneous cooling state for freely cooling dry granular gases) instead of the local equilibrium distribution. A crucial point is that although the granular temperature T is not a conserved field (due to the inelastic character of the collisions), it is assumed that T can still be considered as a slow field. This assumption has been clearly supported by the good agreement found between granular hydrodynamics and computer simulations in several non-equilibrium situations (Lutsko *et al.* 2002; Dahl *et al.* 2002; Lois *et al.* 2007; Mitrano *et al.* 2011; Chialvo & Sundaresan 2013; Mitrano *et al.* 2014). More details on the application of the Chapman–Enskog method to dry (no gas phase) granular mixtures can be found, for example, in Garzó (2019).

Based on the above arguments, in the hydrodynamic regime, the kinetic equation (3.1) admits a normal (or hydrodynamic) solution where all the space and time dependence of f_0 only occurs through a functional dependence on the hydrodynamic fields. As usual (Chapman & Cowling 1970), this functional dependence can be made explicit by assuming small spatial gradients. In this case, f_0 can be written as a series expansion in powers of the spatial gradients of the hydrodynamic fields:

$$f_0 = f_0^{(0)} + f_0^{(1)} + \dots, \quad (5.1)$$

where the approximation $f_0^{(k)}$ is of order k in the spatial gradients. In addition, in the presence of the gravitational force, it is necessary to characterise the magnitude of the force relative to that of the spatial gradients. Here, as in the case of conventional fluid mixtures (Chapman & Cowling 1970), we assume that the magnitude of $\mathbf{g}$ is at least of first order in the perturbation expansion. The implementation of the Chapman–Enskog method to first order in the spatial gradients follows steps similar to those made in the conventional inelastic hard sphere model for dry granular mixtures (Garzó & Dufty 2002; Garzó, Dufty & Hrenya *et al.* 2007a,b).

In contrast to the application of the Chapman–Enskog method for dry granular mixtures, although we are interested here in calculating the diffusion transport coefficients in steady states, the presence of the surrounding molecular gas yields in inhomogeneous states a local energy unbalance between the energy supplied by the bath (or thermostat) and the energy lost by inelastic collisions. This means that we must first consider a time-dependent reference distribution $f^{(0)}(\mathbf{r}, \mathbf{v}, t)$ to obtain the time-dependent linear integral equations that verify the diffusion coefficients. One then assumes stationary conditions and solves (approximately) the above integral equations by considering the so-called first Sonine approximation. In addition, as discussed by Gómez González & Garzó (2022b), the term $\Delta U = U - U_g$ must be considered to be at least of first order in spatial gradients. In this case, the Maxwellian distribution $f_g(\mathbf{r}, \mathbf{v}, t)$ can be written as

$$f_g(\mathbf{v}) = f_g^{(0)}(\mathbf{V}) + f_g^{(1)}(\mathbf{V}) + \dots, \quad (5.2)$$

where

$$f_g^{(0)}(\mathbf{V}) = n_g \left(\frac{m_g}{2\pi T_g} \right)^{d/2} \exp \left(-\frac{m_g V^2}{2T_g} \right), \quad f_g^{(1)}(\mathbf{V}) = -\frac{m_g}{T_g} \mathbf{V} \cdot \Delta U f_g^{(0)}(\mathbf{V}). \quad (5.3)$$

The mathematical steps involved in the determination of the zeroth- and first-order distribution functions are quite similar to those made in previous works on granular mixtures (Garzó & Dufty 2002; Garzó & Montanero 2007). Technical details carried out in this derivation are provided in the supplementary material. In particular, the first-order distribution function $f^{(1)}(\mathbf{r}, \mathbf{v}, t)$ is given by

$$f_0^{(1)}(\mathbf{V}) = \mathcal{A}_0(\mathbf{V}) \cdot \nabla T + \mathcal{B}_0(\mathbf{V}) \cdot \nabla n + \mathcal{C}_0(\mathbf{V}) \cdot \nabla n_0 + \mathcal{D}'_0(\mathbf{V}) \nabla \cdot \mathbf{U} + \mathcal{D}_{0,ij}(\mathbf{V}) \left(\partial_i U_j + \partial_j U_i - \frac{2}{d} \delta_{ij} \nabla \cdot \mathbf{U} \right) + \boldsymbol{\varepsilon}_0(\mathbf{V}) \cdot \Delta U, \quad (5.4)$$

where the unknowns ($\mathcal{A}_0$, $\mathcal{B}_0$, $\mathcal{C}_0$, $\mathcal{D}_{0,ij}$, $\mathcal{D}'_0$, $\boldsymbol{\varepsilon}_0$) are the solutions of a set of coupled linear integral equations displayed in the supplementary material.

5.1. Diffusion transport coefficients

The constitutive equation for the mass flux $\mathbf{j}_0^{(1)}$ is given by (1.1). The diffusion transport coefficients are defined as

$$D_T = -\frac{m_0 T}{d\rho} \int d\mathbf{v} \mathbf{V} \cdot \mathcal{A}_0(\mathbf{V}), \quad (5.5)$$

$$D = -\frac{n}{d} \int d\mathbf{v} \mathbf{V} \cdot \mathcal{B}_0(\mathbf{V}), \quad (5.6)$$

$$D_0 = -\frac{\rho}{dm_0} \int d\mathbf{v} \mathbf{V} \cdot \mathcal{C}_0(\mathbf{V}), \quad (5.7)$$

$$D_0^U = -\frac{m_0}{d} \int d\mathbf{v} \mathbf{V} \cdot \boldsymbol{\varepsilon}_0(\mathbf{V}). \quad (5.8)$$

The procedure for obtaining the expressions of the set of coefficients (D_T , D , D_0 , D_0^U) is described in the the supplementary material and only their final forms are provided in § 6. It is important to recall that the expressions of the diffusion coefficients cannot be analytically obtained for general unsteady conditions since it would require numerically solving a set of coupled differential equations for them. Thus, to reach analytical expressions for these diffusion coefficients, one assumes the validity of the steady-state constraint $\zeta + \zeta_g = 0$ at each point of the system. This allows us to achieve explicit forms for the set (D_T , D , D_0 , D_0^U).

6. Sonine polynomial approximation to the diffusion transport coefficients in steady-state conditions

As mentioned before, the diffusion transport coefficients are given in terms of the solutions of a set of coupled linear integral equations. As usual in kinetic theory of both molecular and granular gases, these integral equations can be approximately solved by considering the leading terms of a Sonine polynomial expansion of the unknowns $\mathcal{A}_0$, $\mathcal{B}_0$, $\mathcal{C}_0$ and $\mathcal{E}_0$. In the case of the mass flux, these quantities are approximated by the polynomials

$$\mathcal{A}_0(V) \rightarrow -f_{0,M} V \frac{\rho}{T n_0 T_0} D_T, \quad \mathcal{B}_0(V) \rightarrow -f_{0,M} V \frac{m_0}{n n_0 T_0} D, \quad (6.1)$$

$$\mathcal{C}_0(V) \rightarrow -f_{0,M} V \frac{m_0^2}{\rho n_0 T_0} D_0, \quad \mathcal{E}_0(V) \rightarrow -f_{0,M} V \frac{D_0^U}{n_0 T_0}. \quad (6.2)$$

To determine D_0 , D , D_T and D_0^U , we substitute first $\mathcal{A}_0$, $\mathcal{B}_0$, $\mathcal{C}_0$ and $\mathcal{E}_0$ by their leading Sonine approximations (6.1) and (6.2) in the corresponding integral equations. Then we multiply these equations by $m_0 V$ and integrate over velocity. Technical details on these calculations are provided in the supplementary material.

6.1. Thermal diffusion coefficient D_T

The thermal diffusion coefficient D_T is given by

$$D_T = \frac{n_0 T}{\rho \nu} D_T^*, \quad D_T^* = \frac{\tau_0 - \frac{m_0}{m} + \chi \frac{\partial \tau_0}{\partial \chi}}{\beta \gamma^* + \nu_D^* + \tilde{\nu}_D \gamma_0^*}, \quad (6.3)$$

where γ_0^* is defined by (4.18), $\tau_0 = T_0/T$ and

$$\beta = (x^{-1} - 3x) \mu^{3/2} \chi^{-1/2}. \quad (6.4)$$

Here, x is given by (4.6) and in (6.3) we have introduced the (reduced) collision frequencies

$$\nu_D^* = \frac{2\pi^{(d-1)/2}}{d\Gamma(\frac{d}{2})} \left(\frac{\sigma'}{\sigma}\right)^{d-1} \mu' \left(\frac{1+\theta_0}{\theta_0}\right)^{1/2} (1+\alpha_0), \quad (6.5)$$

$$\tilde{\nu}_D = \left(\frac{m_0 T_0}{m_g T_g}\right)^{1/2} \mu_{g0} (1+\theta_{0g})^{1/2}, \quad (6.6)$$

where

$$\theta_0 = \frac{m_0 T}{m T_0}, \quad \theta_{0g} = \frac{m_0 T_g}{m_g T_0}. \quad (6.7)$$

While the quantity θ_0 is the ratio of the mean square velocities of the intruders and granular gas particles, the quantity θ_{0g} gives the ratio of the mean square velocities of

the intruders and molecular gas particles. Moreover, the explicit form of the derivative $\partial\tau_0/\partial\chi$ appearing in (6.3) is given in the supplementary material.

In the Brownian limiting case ($m \gg m_g$ and $m_0 \gg m_g$), $\tilde{\nu}_D \rightarrow 1$, $x \rightarrow \chi^{-1/2}$, $\beta \rightarrow 1 - 3\chi^{-1}$ and (6.3) yields

$$D_T^* \rightarrow D_T^{*B} = \frac{\tau_0 - \frac{m_0}{m} + \chi \frac{\partial\tau_0}{\partial\chi}}{\nu_D^* + \gamma_0^* - 2\gamma^*\chi^{-1} - \frac{1}{2}\zeta^*}. \quad (6.8)$$

Upon obtaining (6.8) use has been made of the steady-state condition

$$\chi^{-1}\gamma^* = \gamma^* + \frac{1}{2}\zeta^*. \quad (6.9)$$

The expression (6.8) agrees with previous results derived from the Langevin-like model based on the Fokker–Planck operators (2.17) and (3.12) (Gómez González & Garzó 2022a, 2023).

6.2. Mutual diffusion coefficient D

The mutual diffusion coefficient D is

$$D = \frac{n_0 T}{m_0 \nu} D^*, \quad D^* = \frac{\zeta^* D_T^* - \frac{m_0}{m} + \phi \frac{\partial\tau_0}{\partial\phi}}{\nu_D^* + \tilde{\nu}_D \gamma_0^*}. \quad (6.10)$$

In (6.3) and (6.10), the derivatives $\partial\tau_0/\partial\chi$ and $\partial\tau_0/\partial\phi$ can be seen as a measure of the departure of the perturbed time-dependent state from the steady reference state. Their explicit forms and derivations are provided in the supplementary material.

In the Brownian limit ($m \gg m_g$ and $m_0 \gg m_g$), $\tilde{\nu}_D \rightarrow 1$ and (6.10) reduces to

$$D^* \rightarrow D^{*B} = \frac{\zeta^* D_T^* - \frac{m_0}{m} + \phi \frac{\partial\tau_0}{\partial\phi}}{\nu_D^* + \gamma_0^*}. \quad (6.11)$$

Equation (6.11) is consistent with the results obtained from the Langevin-like model (Gómez González & Garzó 2022a, 2023).

6.3. Tracer diffusion coefficient D_0

The tracer diffusion coefficient D_0 is given by

$$D_0 = \frac{mnT}{m_0^2 \nu} D_0^*, \quad D_0^* = \frac{\tau_0}{\nu_D^* + \tilde{\nu}_D \gamma_0^*}. \quad (6.12)$$

In the Brownian limit,

$$D_0^* \rightarrow D_0^{B*} = \frac{\tau_0}{\nu_D^* + \gamma_0^*}, \quad (6.13)$$

which agrees with previous results (Gómez González & Garzó 2022a, 2023).

6.4. Velocity diffusion coefficient D_0^U

The diffusion coefficient D_0^U is given by

$$D_0^U = m_0 n_0 D_0^{U*}, \quad D_0^{U*} = \frac{\xi_0^* - \xi^*}{\nu_D^* + \tilde{\nu}_D \gamma_0^*}. \quad (6.14)$$

Here,

$$\xi_0^* = \frac{\xi_0}{\rho_0 v} = \mu_0 \theta_0^{-1/2} (1 + \theta_0)^{1/2} \gamma_0^*, \quad (6.15)$$

$$\xi^* = \frac{\xi}{\rho v} = \mu \theta^{-1/2} (1 + \theta)^{1/2} \gamma^*, \quad (6.16)$$

where $\rho_0 = m_0 n_0$ and $\theta = m T_g / (m_g T)$.

In the Brownian limit, $\xi_0^* \rightarrow \gamma_0^*$, $\xi^* \rightarrow \gamma^*$ and $\tilde{v}_D \rightarrow 1$, and (6.14) reduces to

$$D_0^{UB*} = \frac{\gamma_0^* - \gamma^*}{v_D^* + \gamma_0^*}, \quad (6.17)$$

which is consistent with the previous results (Gómez González & Garzó 2022a, 2023) derived from the Langevin-like model.

6.5. Self-diffusion limiting case

Another interesting limiting case corresponds to the self-diffusion problem, namely when the intruders move in a granular gas whose particles are mechanically equivalent to it ($\sigma = \sigma_0$, $m = m_0$, $\alpha = \alpha_0$). In this case, $\xi^* = \xi_0^*$, $\tau_0 = 1$, $\partial_\chi \tau_0 = \partial_\phi \tau_0 = 0$, and so (6.3) and (6.14) yield $D_T = D_0^U = 0$ as expected. Moreover, $D_0^* = -D^*$ and hence the constitutive equation (1.1) for the mass flux becomes

$$\mathbf{j}_0^{(1)} = -\frac{nT}{v} D_{0,self}^* \nabla x_0, \quad (6.18)$$

where $x_0 = n_0/n$ is the concentration (or mole fraction) of the tracer species and the self-diffusion coefficient $D_{0,self}^*$ is

$$D_{0,self}^* = \frac{1}{v_{D,self}^* + \gamma^* \tilde{v}_{D,self}}. \quad (6.19)$$

In (6.16),

$$v_{D,self}^* = \frac{\sqrt{2}\pi^{(d-1)/2}}{d\Gamma\left(\frac{d}{2}\right)} (1 + \alpha), \quad \tilde{v}_{D,self} = \frac{m}{m + m_g} \left(1 + \frac{m_g T}{m T_g}\right)^{1/2}. \quad (6.20)$$

6.6. Some illustrative systems

The expressions of the diffusion transport coefficients D_T , D , D_0 and D_0^U in the steady state are given by (6.3), (6.10), (6.12) and (6.14), respectively. As usual in the study of transport properties in dry granular gases (Brey *et al.* 1998; Garzó & Dufty 1999a), to assess the impact of inelasticity in collisions on diffusion, the diffusion transport coefficients are scaled with respect to their values for elastic collisions ($\alpha = \alpha_0 = 1$). As expected, these scaled diffusion coefficients depend in a complex way on the parameter space (defined by the set (4.14)) of the system. Since the parameter space ξ is large, for the sake of simplicity, henceforth we consider hard spheres ($d = 3$) with $\phi = 0.0052$ (very dilute granular gas), $T_g^* = 10$ and with a common coefficient of restitution $\alpha = \alpha_0$.

Figures 3–6 show the α dependence of the (scaled) coefficients $D_T(\alpha)/D_T(1)$, $D(\alpha)/D(1)$, $D_0(\alpha)/D_0(1)$, and $D_0^U(\alpha)/D_0^U(1)$, respectively. Four different values of the mass ratio m/m_g were considered; in all systems $m_0/m = 8$ and $\sigma_0/\sigma = 2$ ($\sigma_0/\sigma = (m_0/m_g)^{1/3}$). In addition, the results derived from the Brownian limiting case are also

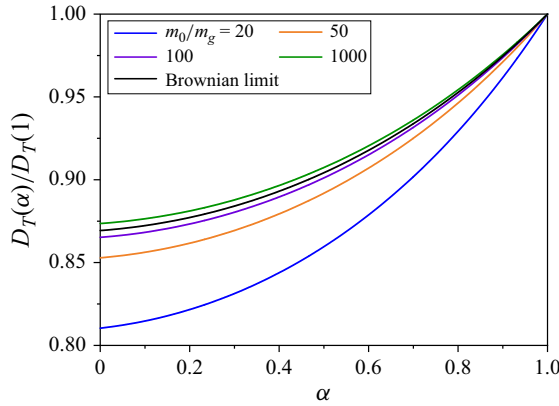


Figure 3. Plot of the (scaled) thermal diffusion coefficient $D_T(\alpha)/D_T(1)$ versus the (common) coefficient of restitution $\alpha = \alpha_0$ for $d = 3$, $\phi = 0.0052$, $T_g^* = 10$ and four different values of the mass ratio m_0/m_g (20, 50, 100 and 1000). In all the curves $m_0/m = 8$, $\sigma_0/\sigma = 2$ and $\sigma_0/\sigma_g = (m_0/m_g)^{1/3}$. The dashed line refers to the expression (6.8) derived in the Brownian limiting case for the ratio $D_T(\alpha)/D_T(1)$.

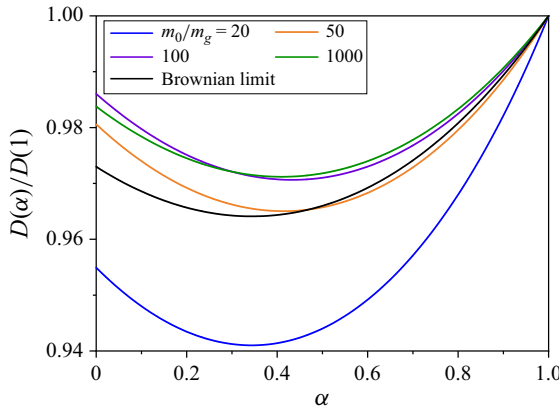


Figure 4. Plot of the (scaled) mutual diffusion coefficient $D(\alpha)/D(1)$ versus the (common) coefficient of restitution $\alpha = \alpha_0$ for $d = 3$, $\phi = 0.0052$, $T_g^* = 10$ and four different values of the mass ratio m_0/m_g (20, 50, 100 and 1000). In all the curves $m_0/m = 8$, $\sigma_0/\sigma = 2$ and $\sigma_0/\sigma_g = (m_0/m_g)^{1/3}$. The dashed line refers to the expression (6.11) derived in the Brownian limiting case for the ratio $D(\alpha)/D(1)$.

shown for comparison. In this case, the diffusion coefficients D_T , D , D_0 and D_0^U are given by (6.8), (6.11), (6.13) and (6.17), respectively. The main objective of figures 3–6 is to show how the combined effect of both inelasticity and the mass ratio m_0/m_g affects the diffusion of intruders in a granular suspension. Additionally, since our results apply to arbitrary values of the mass ratio, we aim to evaluate for practical purposes the conditions under which the diffusion coefficients converge to their values in the so-called Brownian limit case.

We observe that in general the diffusion transport coefficients deviate from their elastic forms, especially for strong inelasticity as expected. However, these deviations are much smaller than those that have been found for dry (no gas phase) granular mixtures (see for example, figures 4, 5 and 6 of Garzó, Murray & Vega Reyes (2013) for $x_0 = 0.2$). This means that the surrounding molecular gas generally hinders the diffusion of tracer particles in a granular gas. While the (scaled) thermal diffusion and tracer diffusion coefficients exhibit a monotonic dependence on the coefficient of restitution

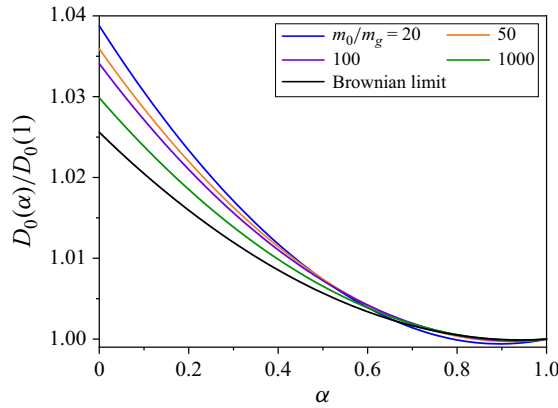


Figure 5. Plot of the (scaled) tracer diffusion coefficient $D_0(\alpha)/D_0(1)$ versus the (common) coefficient of restitution $\alpha = \alpha_0$ for $d = 3$, $\phi = 0.0052$, $T_g^* = 10$ and four different values of the mass ratio m_0/m_g (20, 50, 100 and 1000). In all the curves $m_0/m = 8$, $\sigma_0/\sigma = 2$ and $\sigma_0/\sigma_g = (m_0/m_g)^{1/3}$. The dashed line refers to the expression (6.13) derived in the Brownian limiting case for the ratio $D_0(\alpha)/D_0(1)$.

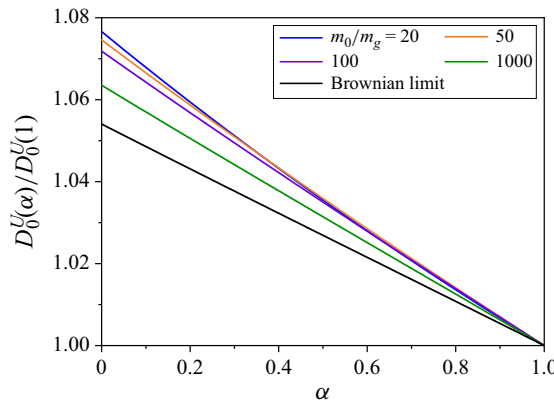


Figure 6. Plot of the (scaled) velocity diffusion coefficient $D_0^U(\alpha)/D_0^U(1)$ versus the (common) coefficient of restitution $\alpha = \alpha_0$ for $d = 3$, $\phi = 0.0052$, $T_g^* = 10$ and four different values of the mass ratio m_0/m_g (20, 50, 100 and 1000). In all the curves $m_0/m = 8$, $\sigma_0/\sigma = 2$ and $\sigma_0/\sigma_g = (m_0/m_g)^{1/3}$. The dashed line refers to the expression (6.17) derived in the Brownian limiting case for the ratio $D_0^U(\alpha)/D_0^U(1)$.

$(D_T(\alpha)/D_T(1) (D_0(\alpha)/D_0(1))$ decreases (increases) with inelasticity) regardless of the mass ratio considered, the (scaled) mutual diffusion coefficient $D(\alpha)/D(1)$ has a non-monotonic dependence on α . This kind of trend is not entirely new: a similar behaviour has already been analysed using a random-walk interpretation in the Fokker–Planck model (Gómez González *et al.* 2023), where the effect was attributed to the competition between two opposite trends: (i) the decrease of the effective mean free path with increasing α , which reduces the persistence of the intruders' trajectories, and (ii) the increase of the collision frequency in the quasielastic regime, which enhances the number of effective steps. The balance between these two competing tendencies explains the emergence of non-monotonicities in the tracer diffusion coefficient. In the present collisional model, the same physical mechanism could explain the observed results.

With respect to the mass ratio, for a fixed value of the (common) coefficient of restitution, it is quite obvious that the (scaled) thermal diffusion coefficient increases

with increasing mass ratio, while the (scaled) tracer diffusion coefficient decreases with increasing mass ratio. The behaviour of the (scaled) mutual diffusion coefficient depends on the range of values of α considered since there are crossings between the different curves. The (scaled) velocity diffusion coefficient has no analogue in the dry granular case. We see that it always increases with increasing inelasticity. Moreover, at a given value of α , it increases with decreasing mass ratio m/m_g . We also observe that the convergence of the curves towards the Brownian limiting case is slower than that found by Gómez González & Garzó (2022b) for monocomponent granular suspensions. In this latter case, the results derived for finite values of m/m_g practically coincide with those obtained in the Brownian limit for values of the mass ratio around 50. Figures 3–6 clearly show that there are still (small) discrepancies between the results derived here and those obtained in the Brownian limit for values of the mass ratio $m_0/m_g = 1000$.

7. An application: segregation of intruders in a granular suspension

As mentioned in § 1, an interesting application of the results displayed in § 6 is the study of the segregation of intruders by thermal diffusion in a granular suspension. Needless to say, thermal diffusion segregation is likely one of the most common phenomena appearing in polydisperse systems. It occurs in a non-convective steady state ($\mathbf{U} = \mathbf{U}_g = \mathbf{0}$) due to the existence of a temperature gradient, which causes the movement of the different species of the mixture. In the steady state, remixing of species generated by diffusion is balanced by segregation caused by temperature differences. To quantify the degree of segregation along the temperature gradient it is usual to introduce the thermal diffusion factor Λ (Kincaid *et al.* 1987). In a steady state without convection and where the mass flux is zero ($\mathbf{j}_0^{(1)} = \mathbf{0}$), the thermal diffusion factor is defined as

$$-\Lambda \frac{\partial \ln T}{\partial z} = \frac{\partial}{\partial z} \ln \left(\frac{n_0}{n} \right), \quad (7.1)$$

where for the sake of simplicity we have assumed that gradients occur only along the z axis. In addition, we also assume that the gravitational field is parallel to the thermal gradient, namely $\mathbf{g} = -g\hat{e}_z$, where $\hat{e}_z$ is the unit vector in the positive direction of the z axis.

We consider a scenario in which the intruders have a larger size than the granular gas particles ($\sigma_0 > \sigma$). Furthermore, as said before, since gravity and the thermal gradient point in the same direction, then the lower plate is hotter than the upper plate ($\partial_z \ln T < 0$). Based on (7.1), when $\Lambda > 0$, intruders rise relative to granular gas particles ($\partial_z \ln(n_0/n) > 0$), leading to an accumulation of tracer particles near the cooler plate. This situation is commonly known as the Brazil nut effect (BNE). Conversely, for $\Lambda < 0$, intruders fall relative to granular gas particles ($\partial_z \ln(n_0/n) < 0$), resulting in an accumulation near the hotter plate. This situation is known as the reverse Brazil nut effect (RBNE). A representative diagram of segregation dynamics is shown in figure 7.

We express the thermal diffusion factor in terms of the diffusion transport coefficients. In the steady state, to first order in spatial gradients, the momentum balance (2.10) reduces to

$$(1 - \varepsilon\kappa^*) + (1 - \varepsilon\bar{\mu}^*) \frac{\partial_z \ln n}{\partial_z \ln T} = -g^*, \quad (7.2)$$

where

$$g^* = \frac{\rho g}{n \partial_z T} < 0 \quad (7.3)$$

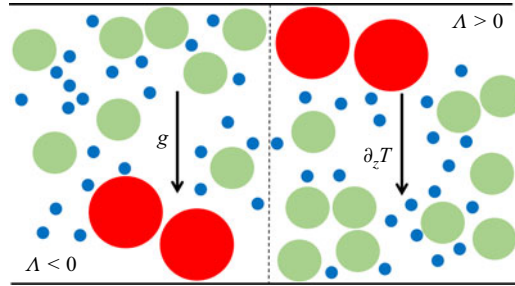


Figure 7. A representative diagram of the BNE ($\Lambda > 0$) and RBNE ($\Lambda < 0$) for a ternary system composed of molecular particles (blue), granular particles (green) and intruders (red).

is a dimensionless parameter measuring the gravity relative to the thermal gradient, κ^* and $\bar{\mu}^*$ are the (reduced) thermal conductivity and diffusive heat conductivity transport coefficients, respectively, and

$$\varepsilon = \frac{(d+2)\sqrt{\pi}}{2^{d+3}(d-1)} \frac{\mu\chi^{-1/2}}{\phi\sqrt{T_g^*}} X(\theta). \quad (7.4)$$

Here, $X(\theta) = \theta^{-1/2}(1+\theta)^{-1/2}$. The explicit forms of κ^* and $\bar{\mu}^*$ are displayed in the supplementary material. Upon obtaining (7.2), use has been made of the result in the leading Sonine approximation (Gómez González & Garzó 2022b):

$$\mathcal{F}^{(1)}[f^{(1)}] \rightarrow \frac{1}{d+2} \frac{\rho\mu\gamma}{n} \kappa_0 X(\theta) (\kappa^* \partial_z \ln T + \bar{\mu}^* \partial_z \ln n), \quad (7.5)$$

where

$$\kappa_0 = \frac{d(d+2)^2 \Gamma\left(\frac{d}{2}\right)}{16(d-1)\pi^{\frac{d-1}{2}}} \sigma^{1-d} \sqrt{\frac{T}{m}} \quad (7.6)$$

is the low-density value of the thermal conductivity for an ordinary gas of hard spheres.

According to (1.1), when $\Delta U = 0$, the condition $j_{0,z}^{(1)} = 0$ yields the relation

$$-D_0^* \partial_z \ln n_0 = D^* \partial_z \ln n + D_T^* \partial_z \ln T, \quad (7.7)$$

where D_T^* , D^* and D_0^* are given by (6.3), (6.10) and (6.12), respectively. From (7.2) and (7.7), the thermal diffusion factor can be written as

$$\Lambda = \frac{\partial_z \ln n}{\partial_z \ln T} - \frac{\partial_z \ln n_0}{\partial_z \ln T} = \frac{D_T^* + (D_0^* + D^*) (\varepsilon \kappa^* - 1 - g^*) (1 - \varepsilon \bar{\mu}^*)^{-1}}{D_0^*}. \quad (7.8)$$

In the Brownian limit ($m/m_g \rightarrow \infty$), $\mu \rightarrow 1$ and $\theta \rightarrow \infty$, so that $\varepsilon \rightarrow 0$. In this limiting case, Λ reduces to

$$\Lambda = \frac{D_T^* - (D_0^* + D^*) (1 + g^*)}{D_0^*}. \quad (7.9)$$

Equation (7.9) agrees with previous results derived for the segregation of massive intruders in a granular suspension (Gómez González & Garzó 2022a).

Since (6.12) clearly shows that $D_0^* > 0$, then the curves delineating the regimes between the segregation towards the cold and the hot wall (BNE/RBNE transition) are determined

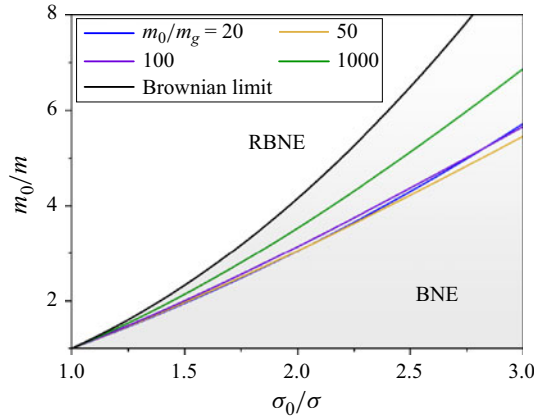


Figure 8. Plot of the marginal segregation curve ($\Lambda = 0$) for a (common) coefficient of restitution $\alpha = \alpha_0 = 0.7$ and $|g^*| \rightarrow 0$. The parameters used are: $d = 3$, $\phi = 0.0052$, $T_g^* = 10$ and four different values of the mass ratio m_0/m_g (20, 50, 100 and 1000). The points below the curve correspond to $\Lambda > 0$ (BNE), while the points above the curve correspond to $\Lambda < 0$ (RBNE). The dashed line is the result obtained in the Brownian limiting case.

from the condition

$$(1 - \varepsilon \bar{\mu}^*) D_T^* = - (D_0^* + D^*) (\varepsilon \kappa^* - 1 - g^*). \quad (7.10)$$

7.1. Mechanically equivalent particles

In this scenario, $D_T^* = 0$ and $D^* = -D_0^*$; thus, (7.10) is valid for any values of the coefficients of restitution, masses and diameters. Consequently, as in the Brownian limit (Gómez González & Garzó 2023), no segregation occurs in the system.

7.2. Different mechanical properties

7.2.1. Absence of gravity ($|g^*| \rightarrow 0$)

Let us first consider a scenario where gravitational effects are negligible ($|g^*| \rightarrow 0$). Under this assumption, the condition $\Lambda = 0$ reads

$$(1 - \varepsilon \bar{\mu}^*) D_T^* = - (D_0^* + D^*) (\varepsilon \kappa^* - 1). \quad (7.11)$$

Figure 8 shows the BNE/RBNE phase diagram for a three-dimensional system ($d = 3$) with a common coefficient of restitution $\alpha = \alpha_0 = 0.7$, $T_g^* = 10$ and five distinct values of the mass ratio m_0/m_g (20, 50, 100, 1000, and the Brownian limit $m_0/m_g \rightarrow \infty$). At first glance, we notice a quantitative discrepancy with the diagrams shown in Gómez González & Garzó (2023) for $|g^*| \rightarrow 0$. These discrepancies were expected since, in that work, segregation was calculated for moderate densities, unlike here ($\phi = 0.0052$). Also, when using the Fokker–Planck approach (3.12) for the operator $J_{0g}[f_0, f_g]$, we see that the expression (4.18) of γ_0^* employed here slightly differs from the expression of $\gamma_{0,FP}^*$ defined by Gómez González & Garzó (2023). Specifically, $\gamma_{0,FP}^* = (\sigma_0/\sigma)(\bar{\sigma}/\bar{\sigma}_0)^{d-1}\gamma_0^*$. This discrepancy is because $\gamma_{0,FP}^*$ was obtained from the granular literature using phenomenological arguments.

However, what is surprising is the complete reversal we see in the diagram when we observe the RBNE effect as we increase m_0/m , contrary to what is observed in Gómez González & Garzó (2023). This requires a more subtle explanation. When calculating the diagrams in the case of an effective model where the thermostat is modelled by a Fokker–Planck equation, the thermostat intensity is regulated only by the (dimensionless) bath

temperature T_g^* . If this parameter is kept constant, as in the figures 11 and 12 reported by Gómez González & Garzó (2023), the thermostat does not change when the mass (m_0/m) or size (σ_0/σ) ratios are modified. However, in our case, we modify m_0/m (or σ_0/σ) keeping m_0/m_g constant for each particular curve. Thus, by modifying the mass ratio m_0/m for a particular value of m_0/m_g , the relative mass between the granular and molecular gas changes. Therefore, the effect of the collisions between the grains and the particles of the molecular gas will be different. Concretely, if we increase m_0/m without changing m_0/m_g , the molecular gas will have a mass increasingly similar to that of the grains. Consequently, the thermalising effect of the molecular gas that compensates for the effect of inelasticity will be more effective, causing the temperature of the granular gas to be higher and thus tend to increase with respect to that of the intruders (RBNE). We observe the same when we increase the size ratio (σ_0/σ). In this case, for a given m_0/m_g (or equivalently σ_0/σ_g), as the size of the intruders increases, the grains will have a size increasingly similar to the particles of the surrounding molecular gas; thus the effective area of the grain in a collision decreases, and, with it, the number of collisions. Therefore, the grain will have less effective thermalisation and will move to the cold zone (BNE).

On the other hand, as we increase m_0/m_g , we see that the transition to the RBNE phase occurs earlier. This is because, by increasing the mass more rapidly than the size ($m_0/m_g = (\sigma_0/\sigma_g)^d$), the effect of intruder–molecular gas collisions in the motion of intruders becomes less effective. In the curves where the mass ratios m_0/m_g are close to each other, the relative size also plays an important role, and crossings can be observed, as was the case in figure 4.

Moreover, for elastic collisions ($\alpha = \alpha_0 = 1$), the segregation criterion notably deviates from the classical result obtained for molecular mixtures of hard spheres by Kincaid *et al.* (1987), where, in the first Sonine approximation, the condition $\Lambda = 0$ yields the simple segregation criterion $m_0/m = 1$. For the present system (intruders moving in a granular gas immersed into a molecular gas), our analysis reveals that there is no segregation for the remaining parameters considered in figure 8.

7.2.2. Thermalised systems ($\partial_z T \rightarrow 0$)

Let us explore a scenario where gravity is the main factor influencing segregation dynamics. In this situation, $|g^*| \rightarrow \infty$, which makes the temperature gradient negligible ($\partial_y T \rightarrow 0$), and the condition $\Lambda = 0$ leads to the relation

$$D_0^* + D^* = 0. \quad (7.12)$$

Figure 9 shows the case $|g^*| \rightarrow \infty$ with the same parameters as figure 8. In this strong-gravity case, the explanation is simpler. Gravity is much stronger than both the energy coming from the molecular gas and the energy lost in inelastic collisions, as seen in granular suspensions and driven granular gases (Garzó 2008; Gómez González & Garzó 2023; Gómez González *et al.* 2024). Because of this, all particles fall to the bottom plate. Heavier intruders are harder to lift, so they move downward (RBNE). However, if the intruders are larger but keep the same mass, smaller particles hit more often. These collisions push the smaller particles below the intruders, lifting them and creating a buoyancy effect (BNE)

For elastic collisions ($\alpha = \alpha_0 = 1$), the cooling rate vanishes ($\zeta^* = 0$), and there is equipartition of energy ($\tau_0 = 1$). Thus, it is straightforward to verify from (6.10) and (6.12) that the segregation criterion simplifies to $m_0/m = 1$. This result aligns with previous findings for dry granular gases (i.e. in the absence of an interstitial gas phase) (Brey *et al.* 2005; Garzó 2006, 2011), as well as with those obtained in the coarse-grained approach by using the Fokker–Planck equation (Gómez González & Garzó 2023).

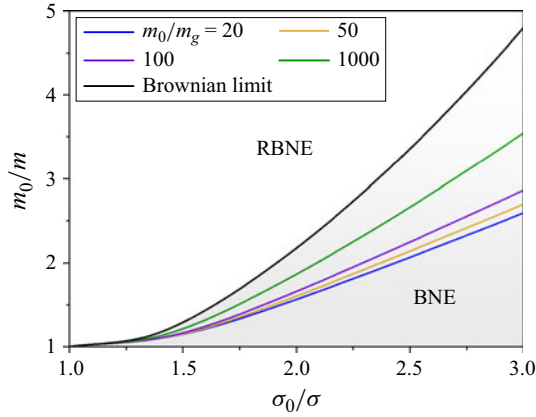


Figure 9. Plot of the marginal segregation curve ($\Lambda = 0$) for a (common) coefficient of restitution $\alpha = \alpha_0 = 0.7$ and $|g^*| \rightarrow \infty$. The parameters used are: $d = 3$, $\phi = 0.0052$, $T_g^* = 10$ and four different values of the mass ratio m_0/m_g (20, 50, 100 and 1000). The points below the curve correspond to $\Lambda > 0$ (BNE), while the points above the curve correspond to $\Lambda < 0$ (RBNE). The dashed line is the result obtained by using the Fokker–Planck approach (2.17) to the operator $J_g[f, f_g]$.

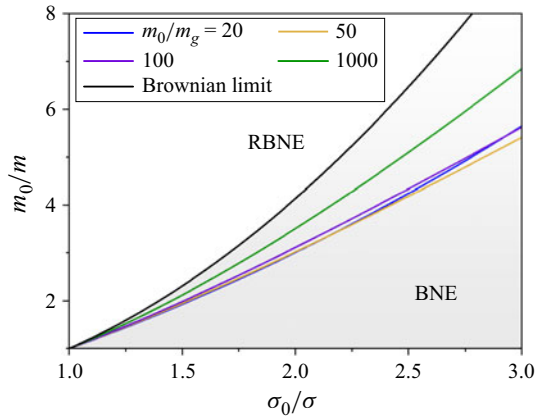


Figure 10. Plot of the marginal segregation curve ($\Lambda = 0$) for a (common) coefficient of restitution $\alpha = \alpha_0 = 0.7$ and $|g^*| = 1$. The parameters used are: $d = 3$, $\phi = 0.0052$, $T_g^* = 10$ and four different values of the mass ratio m_0/m_g (20, 50, 100 and 1000). The points below the curve correspond to $\Lambda > 0$ (BNE), while the points above the curve correspond to $\Lambda < 0$ (RBNE). The dashed line is the result obtained by using the Fokker–Planck approach (2.17) to the operator $J_g[f, f_g]$.

7.2.3. General case

As a final situation, we consider the general case where the effects of the temperature gradient and gravity are comparable. To exemplify this, figure 10 shows the marginal segregation curve for a reduced gravity $|g^*| = 1$ and for the same systems as depicted in figures 8 and 9.

The main point is that, unlike in dry granular mixtures and granular suspensions (Garzó 2019; Gómez González & Garzó 2023; Gómez González *et al.* 2024), gravity has a weaker effect than the thermal gradient, keeping the BNE/RBNE transition quite similar to that shown in figure 8. The explanation can be the same as in the case without gravity since, when increasing m_0/m , the relative mass of the grains compared with molecular particles decreases. As a result, the thermalisation due to the surrounding fluid does not

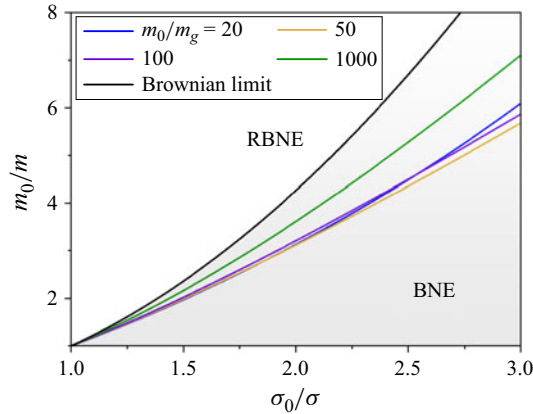


Figure 11. Plot of the marginal segregation curve ($\Lambda = 0$) for a (common) coefficient of restitution $\alpha = \alpha_0 = 1$ and $|g^*| = 1$. The parameters used are: $d = 3$, $\phi = 0.0052$, $T_g^* = 10$ and four different values of the mass ratio m_0/m_g (20, 50, 100 and 1000). The points below the curve correspond to $\Lambda > 0$ (BNE), while the points above the curve correspond to $\Lambda < 0$ (RBNE). The dashed line is the result obtained by using the Fokker–Planck approach (2.17) to the operator $J_g[f, f_g]$.

remain constant, making thermal effects more pronounced even when gravity is present. To complement the phase diagram shown in figure 10 for inelastic collisions, in figure 11 we plot the marginal segregation curve ($\Lambda = 0$) for the same values of the mass ratio m_0/m_g as in figure 10 but for elastic collisions. It is quite apparent from the comparison between figures 10 and 11 that the inelasticity of collisions plays a secondary role in the segregation behaviour in this case since we observe practically no differences between both figures.

8. Summary and concluding remarks

This paper aims to determine the diffusion transport coefficients for tracer (or intruder) particles within a granular gas modelled as a gas of inelastic hard spheres. Intruders and granular gas are immersed in a bath of elastic hard spheres (molecular gas). We examine scenarios where the granular particles are sufficiently dilute, ensuring the molecular gas remains unaffected and serves as a thermostat at a given temperature T_g . Unlike other suspension models, which consider an effective fluid–solid force, our model accounts for both inelastic collisions between the tracer and granular particles, as well as among the granular particles themselves. Additionally, it takes into account the elastic collisions between the grains and molecular gas particles, as well as between the intruders and molecular gas. We consider the low-density regime for the suspended solid particles and, hence, the velocity distribution functions $f(\mathbf{r}, \mathbf{v}, t)$ and $f_0(\mathbf{r}, \mathbf{v}, t)$ for grains and intruders, respectively, obey the (inelastic) Boltzmann equations.

To ensure a consistent theoretical framework, we first analyse homogeneous reference states as a basis for applying the Chapman–Enskog method and obtaining then the diffusion transport coefficients. In the homogeneous state, we present new results for the temperature ratio $\chi_0 = T_0/T_g$ and the kurtosis c_0 associated with the intruders. As in the case of the granular gas (Gómez González & Garzó 2022b), we find that the tracer temperature ratio χ_0 shows larger deviations from unity as the mass ratio m_0/m_g decreases. Regarding the kurtosis c_0 (which measures the departure of the tracer distribution from its Maxwellian form), as for the granular gas (Gómez González & Garzó 2022b), our results clearly show that this quantity remains small enough to validate the use of the Maxwellian approximation for the velocity distributions.

We compared the theoretical results derived in the HSS for χ_0 and c_0 with DSMC data, and the agreement is remarkable. This justifies the use of the Maxwellian distribution (4.12) to achieve accurate estimates of the cooling rates ζ_0 and ζ_{0g} . We also observe the convergence of the results in the Brownian limit ($m/m_g \rightarrow \infty$ and $m_0/m_g \rightarrow \infty$) with those obtained in previous works (Gómez González & Garzó 2019, 2023) by using the Fokker–Planck approach.

Once the homogeneous state is characterised, the corresponding set of kinetic equations for the mixture were addressed using the Chapman–Enskog method (Chapman & Cowling 1970), up to first order in spatial gradients. From this solution, the four diffusion transport coefficients are derived by considering the leading terms in a Sonine polynomial expansion of the first-order distribution function. In dimensionless form, these diffusion coefficients are given in terms of eight dimensionless parameters: the diameter ratios σ/σ_g and σ_0/σ_g , the mass ratios m/m_g and m_0/m_g , the coefficients of restitution α and α_0 , the (reduced) bath temperature T_g^* and the volume fraction ϕ (which is considered to be quite small since we are considering a very dilute granular gas). Compared with previous attempts to obtain tracer diffusion coefficients in granular suspensions (see e.g. Gómez González & Garzó 2023) by starting from a coarse-grained approach, our results provide a more general description of the dependence of diffusion on mass (m/m_g and m_0/m_g) and diameter (σ/σ_g and σ_0/σ_g) ratios. As expected, the expressions for the tracer diffusion coefficients agree with previous results derived in the Brownian limit from the Fokker–Planck operator (Gómez González & Garzó 2023). Thus, with respect to previous attempts for obtaining tracer diffusion coefficients in granular suspensions (Gómez González & Garzó 2023) by starting from a coarse-grained approach, our results provide a more general description of the dependence of the diffusion on the mass (m/m_g and m_0/m_g) and diameter (σ/σ_g and σ_0/σ_g) ratios. In particular, as expected, the expressions for the tracer diffusion coefficients agree with previous results derived in the Brownian limit from the Fokker–Planck operator (Gómez González & Garzó 2023). In this context, the present results encompass the previous ones as a special case, thus covering interesting physical situations that had not previously been analysed from a theoretical perspective.

In general, the diffusion coefficients exhibit significant deviations from their elastic counterparts, particularly under strong inelasticity. A key result is that convergence to the predictions of the effective model is achieved only at large mass ratios $m_0/m_g \approx 1000$, which contrasts with the convergence observed in a monocomponent granular suspension where agreement was found at $m/m_g = 50$ (Gómez González & Garzó 2022a). This emphasises the ability of the model to capture more realistic scenarios beyond the scope of Fokker–Planck-based models.

As an application, we investigate segregation driven by both thermal gradients and gravity. We find that increasing the mass ratio m_0/m_g tends to push the intruders towards the bottom plate (RBNE), regardless of the gravitational strength. This behaviour contrasts with the results derived from the Fokker–Planck model (Gómez González & Garzó 2023). In that model, the thermostat effect remains constant (set by T_g^*), whereas in our model it depends on m_0/m_g , resulting in different thermalisation dynamics. The main novelty of the study reported here, compared with previous works on dry granular mixtures (see e.g. Garzó 2008, 2011), is the analysis of the role of m_0/m_g at fixed α . Unlike the dry case, where inelasticity strongly affects BNE/RBNE transitions due to energy non-equipartition, here its impact is reduced because the molecular gas injects energy homogeneously, compensating collisional dissipation and effectively suppressing partial temperature differences through thermalisation.

The results reported in this work suggest a framework to study experimentally the influence of an interstitial gas on segregation dynamics. In this context, a feasible

experiment can be designed following the approach discussed in Gómez González *et al.* (2023). A suitable system to represent the low-Reynolds-number regime, in which the effect of granular collisions is comparable to the effect of the thermal bath, can be realised by immersing gold grains in hydrogen gas at low pressure. In this set-up, the molecular gas provides a homogeneous thermal bath with appropriate viscosity η_g , such that the Reynolds numbers remain very small ($Re \sim 10^{-4}$) and the Stokes number, T_g^* , as well as γ/ν , are all close to unity. This ensures that the molecular gas effectively thermalises the granular particles, while the effect of granular collisions remains significant.

Once the particles composing the system are selected, an experimental set-up to investigate segregation of intruders in a granular gas immersed in an interstitial fluid can be designed based on previous studies (Möbius *et al.* 2001; Naylor, Swift & King 2003; Sánchez, Swift, & King 2004; Wylie *et al.* 2008; Clement *et al.* 2010; Pastenes, Géminard & Melo 2014). The set-up can consist of a transparent container with a porous base to allow fluid flow while maintaining particle collisions, filled with small granular particles and a single intruder whose mass and size can be adjusted. The interstitial fluid, either a gas or a liquid, occupies the voids between particles, enabling the study of drag and thermalisation effects. The container is subjected to controlled vertical or horizontal vibrations with adjustable amplitude, and the position of the intruder is tracked using high-speed imaging. By varying the intruder's mass and size and the properties of the fluid, this set-up allows for systematic measurement of segregation phenomena, including the influence of mass ratio and interstitial fluid on the BNE/RBNE transition.

Finally, it is important to remark that to facilitate the solution of the integral equations verifying the mixture, we have considered the tracer limit where one of the species is present at a negligible concentration. A possible extension of this work is to generalise diffusion to a binary mixture with arbitrary concentrations. In addition, it may be interesting to study the role of density in the diffusion and segregation diagrams using the Enskog kinetic equation which applies to moderate densities. Moreover, revisiting the problem within a simplified random-walk framework could shed light on the underlying mechanisms responsible for the observed non-monotonicities and the crossing of curves in the tracer diffusion coefficient. These studies will be developed in future projects.

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