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# Particle collision modeling – A review

C.J. Meyer [a], D.A. Deglon [b,*]

[a] Centre for Research in Computational and Applied Mechanics, University of Cape Town, Rondebosch, South Africa
[b] Centre for Minerals Research, University of Cape Town, Rondebosch, South Africa

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ABSTRACT

Over the past 100 years particle collision models for a range of particle inertias and carrier fluid flow conditions have been developed. Models for perikinetic and orthokinetic collisions for simple, laminar shear flows as well as collisions associated with differential sedimentation are well documented. Collision models developed for turbulent flow conditions are demarcated on the one side with the model of Saffman and Turner (1956) associated with particles exhibiting zero inertia and on the other side with the model of Abrahamson (1975) for particle velocities that are completely decorrelated from the carrier fluid velocities. Various attempts have been made to develop universal collision models that span the entire range of inertias in a turbulent flow field. It is a well-accepted fact that models based on a cylindrical as opposed to a spherical formulation are erroneous. Furthermore, the collision frequency of particles exhibiting identical inertias are not negligible. Particles exhibiting relaxation times close to the Kolmogorov time scale of the turbulent flow are subject to preferential concentration that could increase the collision frequency by up to two orders of magnitude. In recent years the direct numerical simulation (DNS) of colliding particles in a turbulent flow field have been preferred as a means to secure the collision data on which the collision models are based. The primary advantage of the numerical treatment is better control over flow and particle variables as well as more accurate collision statistics. However, a numerical treatment places a severe restriction on the magnitude of the turbulent flow Reynolds number. The future development of more comprehensive and accurate collision models will most likely keep pace with the growth in computational resources.



**Contents**



## 1. Introduction

Particle collision constitutes an important sub-process in a wide range of natural occurring as well as industrial processes where the agglomeration and/or breakup of particles is of importance. These

* Corresponding author.
*E-mail addresses:* cjmeyer@sun.ac.za (C.J. Meyer), David.Deglon@uct.ac.za (D.A. Deglon).

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processes involve a continuous phase (liquid or gas) and one or more dispersed phases (solid and/or liquid and/or gas). Where the continuous phase is a liquid, as is common in mineral processing, the dispersed phase/s may be a solid (particle) and/or a liquid (droplet) and/or a gas (bubble).

Natural processes characterized by particle collision range from planetary formation from protoplanetary nebula Champney et al. (1995) to the formation of rain drops in clouds Pinsky et al. (2000).

Particle collision is relevant to many industrial processes. Examples include, amongst others, the aggregation of solid particles in flocculation/sedimentation Balthasar et al. (2002), the rate of coalescence of droplets/bubbles in liquid and gas dispersions Kamp et al. (2001), Narsimhan (2004), the interaction between particles and bubbles in froth flotation Schubert (1999), Bloom and Heindel (2002), Deglon (2005), the secondary nucleation of crystals in crystallization ten Cate et al. (2001) and soot formation in furnaces Balthasar et al. (2002).

Particle collision is particularly relevant to mineral processing as turbulent multiphase systems are common and many subprocesses are controlled/influenced by turbulent collision. Most of the examples quoted previously are relevant to common unit operations in mineral processing (e.g. thickening, solvent extraction, froth flotation, crystallization).

This paper is an effort to present the various approaches used to model dispersed phase collision as it pertains to industrial processes. The discussion commences with the presentation of the general collision modeling approach with special attention being given to the definition and interpretation of concepts central to the modeling approach. This is followed by a listing and discussion of appropriate collision models with emphasis being placed on formulations for collision frequency. The expressions for collision frequency are augmented, in the case of turbulent flow, with formulations of velocity fluctuations.

## 2. Collision modeling

Smoluchowski (1917), using a population balance approach, formulated the following expression to quantify the agglomeration of particles due to fluid agitation

$$\frac{\partial N(r_i,t)}{\partial t} = \frac{1}{2}\sum_{j=1}^{i-1}\beta(r_i-r_j,r_j)N(r_i-r_j,t)N(r_j,t) - \sum_{j=1}^{\infty}\beta(r_i,r_j)N(r_i,t)N(r_j,t) \quad (1)$$

where $N$ is the number density and $\beta$ the collision kernel or frequency (number of collisions per unit volume and time). The latter describes the rate at which particles of size $(r_i - r_j)$ collide with particles of size $r_j$ to form particles of size $r_i$ (first term) and also how particles of size $r_i$ collide with particles of size $r_j$ to reduce the number of particles with size $r_i$ (second term).

Analytical solutions to Eq. (1) exist where the collision kernel takes on a simple form, such as a constant value for instance. However, in most practical applications the kernel takes on a significantly more complex form that depends, to a large extent, on the flow and particle kinematics.

The primary collision mechanisms are listed in Table 1 where the Stokes number, $St$, contrasts the particle relaxation time, $\tau_i$, with that of the smallest scales of fluid motion, $\tau_\eta$. In the case of fully developed turbulent flow the latter will constitute the Kolmogorov micro time scale, $\sqrt{\nu/\epsilon}$ where $\nu$ is the fluid kinematic viscosity and $\epsilon$ the dissipation rate of turbulent kinetic energy per unit mass.

In their 1955 publication, Telford et al. (1955) make mention of a controversy raised by the work of Defant (1905) who observed that rain droplets seemed to be grouped about masses m, 2 m, 4 m, 8 m, etc. In other words, droplets of a particular size preferentially combine with droplets of a similar size. Based on their work, Telford et al. (1955) attributed this phenomena to droplet wake effects.

Saffman and Turner (1956) noted that the motion of smaller droplets are strongly influenced by the presence of larger droplets which in turn significantly affects the rate of coagulation of smaller droplets with larger droplets. The effect of this interference is accounted for through the introduction of a collision efficiency, $\alpha$.

Eq. (1) can thus be cast into the general form

$$\frac{\partial N_k}{\partial t} = \alpha \cdot \beta \cdot N_i \cdot N_j \quad (2)$$

where $N_k$, $N_i$ and $N_j$ are the number densities of the aggregate and two dispersed particle sizes respectively.

However, in their review of flocculation modeling, Thomas et al. (1999) refer to an alternative interpretation of $\alpha$ where it is said to account for inaccuracies associated with the collision kernel itself. In fact, Thomas et al. (1999) state that hydrodynamic interference can be more accurately interpreted as modifications to the collision kernel rather than a collision efficiency and that the latter is most notably affected by short-range forces i.e electrostatic repulsion and van der Waals attraction.

### 2.1. Perikinetic

Smoluchowski (1917) was able to demonstrate that for Brownian motion, the collision frequency can be expressed by Eq. (26) (see Table 2), where $\kappa_B$ is the Boltzmann constant, $T$ the absolute temperature and $\mu$ the fluid viscosity. This expression has the following assumptions as basis

1. The collision efficiency is unity.
2. The fluid motion is laminar.

**Table 1**
Modes of collision.

| Mechanism | Description | Continuous phase flow regime | Scale and flow regime of dispersed phase |
|---|---|---|---|
| Brownian motion (perikinetic) | Particle collision due to random Brownian motion of particles | Laminar | Particles are small, less than 1 μm |
| Shear (orthokinetic) | Particles follow streamlines and collide due to different positions within shear flow field | Laminar and turbulent | Various length scales; $St \leqslant 1$ |
| Differential sedimentation | Particles of different sizes exhibit different settling velocities leading to collisions | Laminar | Various length scales; Various particle relaxation times |
| Accelerative – correlated | Particles deviate from streamlines and collide. Particle and carrier fluid velocities are correlated or partly correlated | Turbulent | Intermediate particle sizes; Various particle relaxation times |
| Accelerative – independent | Particles are thrown randomly from eddy to eddy and collide. Particle and carrier fluid velocities are uncorrelated | Highly turbulent | Particles are larger than viscous dissipation eddies; $St \geqslant 10$ |

**Table 2**
Collision frequency.

| Reference | Mode | Frequency | |
|---|---|---|---|
| Smoluchowski (1917) | Perikinetic | $\beta = \left(\frac{2k_BT}{3\mu}\right)\left(\frac{1}{r_i}+\frac{1}{r_j}\right)(r_i+r_j)$ | (26) |
| | Orthokinetic | $\beta = \left(\frac{4}{3}\right) \left\vert \frac{dU_x}{dy} \right\vert (r_i+r_j)^3$ | (27) |
| Camp and Stein (1943) | Orthokinetic | $\beta = \left(\frac{4}{3}\right)G(r_i+r_j)^3$<br>$G = \sqrt{(\Phi/\mu)}$, laminar<br>$G = \sqrt{(\overline{\Phi}/\mu)}$, turbulent | (28) |
| | Differential sedimentation | $\beta = \left(\frac{2g\pi}{9\mu}\right)(\rho_{i,j}-\rho)(r_i+r_j)^3\lvert r_i-r_j\rvert$ | (29) |
| Saffman and Turner (1956) | Orthokinetic | $\beta = \sqrt{\frac{8\pi}{15}}(r_i+r_j)^3\left(\frac{\epsilon}{\nu}\right)^{1/2}$ | (30) |
| | Accelerative – correlated | $\beta = \sqrt{8\pi}(r_i+r_j)^2\left[\left(1-\frac{\rho}{\rho_{i,j}}\right)^2(\tau_i-\tau_j)^2\langle\left(\frac{Du_x}{Dt}\right)^2\rangle + \cdots\right.$<br>$\left. +\frac{1}{3}(1-\frac{\rho}{\rho_p})^2(\tau_i-\tau_j)^2g^2 + \frac{\epsilon}{9\nu}(r_i+r_j)^2\right]^{1/2}$ | (31) |
| Argaman and Kaufman (1968) | Orthokinetic | $\beta = 4\pi K_s r_i^3\langle u_x^2\rangle, \langle u_x^2\rangle = K_pG$ | (32) |
| Abrahamson (1975) | Accelerative – independent | $\beta = (r_i+r_j)^2\left[\sqrt{8\pi}\sqrt{\langle v_i^2\rangle+\langle v_j^2\rangle}\exp\left(-\frac{(w_{tj}-w_{ti})^2}{2(\langle v_{,i}^2\rangle+\langle v_{,j}^2\rangle)}\right)+\cdots\right.$<br>$\left. +\pi\frac{(w_{tj}-w_{ti})^2+\langle v_i^2\rangle+\langle v_j^2\rangle}{w_{tj}-w_{ti}} erf\left(\frac{w_{tj}-w_{ti}}{\sqrt{2(\langle v_i^2\rangle+\langle v_j^2\rangle)}}\right)\right]$<br>$\langle v_i^2\rangle = \frac{\langle u^2\rangle}{1+1.5\tau_i\epsilon/\langle u^2\rangle}, \quad \langle v_j^2\rangle = \frac{\langle u^2\rangle}{1+1.5\tau_j\epsilon/\langle u^2\rangle}$ | (33) |
| Delichatsios and Probstein (1975) | Orthokinetic | $\beta = \frac{\pi}{2}d^2w'_x$<br>$w'_x = \left(\frac{\epsilon}{15\nu}\right)^{1/2}d, \quad d < \eta$<br>$w'_x = 1.37(\epsilon d)^{1/3}, \quad d > \eta$<br>$w'_x = 1.37(\epsilon L)^{1/3}, \quad d \sim L$ | (34) |
| Williams and Crane (1983) | Accelerative – correlated | $\beta = \sqrt{\frac{8\pi}{3}}(r_i+r_j)^2\sqrt{3\langle w_x^2\rangle}$<br>$\frac{\langle w_x^2\rangle}{\langle u_x^2\rangle} = \frac{\gamma(\theta_i-\theta_j)^2}{(\gamma-1)(\theta_i+\theta_j)}\left(\frac{1}{(1+\theta_i)(1+\theta_j)}-\frac{1}{(1+\gamma\theta_i)(1+\gamma\theta_j)}\right), \theta_i \text{ and } \theta_j \ll 1$<br>$\frac{\langle w_x^2\rangle}{\langle u_x^2\rangle} = \frac{1}{1+\theta_i}+\frac{1}{1+\theta_j}-\frac{4}{\theta_i+\theta_j+\theta_i\theta_j\sqrt{\frac{1}{1+\theta_i}+\frac{1}{1+\theta_j}}}, \theta_i \text{ or } \theta_j \gg 1$<br>$\frac{\langle w_x^2\rangle}{\langle u_x^2\rangle} = \frac{(\theta_i+\theta_j)^2-4\theta_i\theta_j\sqrt{\frac{1+\theta_i+\theta_j}{(1+\theta_i)(1+\theta_j)}}}{(\theta_i+\theta_j)(1+\theta_i)(1+\theta_j)}$, universal | (35) |
| Yuu (1984) | Accelerative – correlated | $\beta = \sqrt{\frac{8\pi}{3}}(r_i+r_j)^2\sqrt{\langle \boldsymbol{w}_{\boldsymbol{accel}}^2\rangle+\langle \boldsymbol{w}_{\boldsymbol{shear}}^2\rangle}$<br>$\langle \boldsymbol{w}_{\boldsymbol{accel}}^2\rangle = (A_i-2B+A_j)\langle \boldsymbol{u}^2\rangle$<br>$\langle \boldsymbol{w}_{\boldsymbol{shear}}^2\rangle = (A_ir_i^2+2Br_ir_j+A_jr_j^2)\left(\frac{\epsilon}{3\nu}\right)$<br>$A_i = \frac{a_iT_L+b^2}{a_iT_L+1}, A_j = \frac{a_jT_L+b^2}{a_jT_L+1}$<br>$B = \frac{C}{(a_i+a_j)(1-a_i^2T_L^2)(1-a_j^2T_L^2)}$<br>$C = a_ia_jT_L(2-(a_i+a_j)T_L-(a_i^2+a_j^2)T_L^2+a_ia_j(a_i+a_j)T_L^3)+\cdots$<br>$+b(a_i-a_j)^2T_L(1-(a_i+a_j)T_L+a_ia_jT_L^2)+\cdots$<br>$+b^2((a_i+a_j)-(a_i^2+a_j^2)T_L-a_ia_j(a_i+a_j)T_L^2+2a_i^2a_j^2T_L^3)$ | (36) |
| Hu and Mei (1997) | Accelerative – correlated | $\beta = 2\pi(r_i+r_j)^2\left(\frac{2}{15\pi}(r_i+r_j)^2\frac{\epsilon}{\nu}+\frac{2}{\pi}\langle\left(\frac{Du_x}{Dt}\right)^2\rangle(\tau_i+\tau_j)^2+\cdots\right.$<br>$\left.+\frac{4\tau_i\tau_j}{\pi}\langle\left(\frac{Du_x}{Dt}\right)^2\rangle\frac{(r_i+r_j)^2}{\lambda_f^2}\right)^{1/2}$ | (37) |
| Kramer and Clark (1997) | Orthokinetic | $\beta = \frac{4\pi}{3}\lvert a'_{max}\rvert(r_i+r_j)^3$ | (38) |
| Kruis and Kusters (1997) | Accelerative – correlated | $\beta = \sqrt{\frac{8\pi}{3}}(r_i+r_j)^2\sqrt{\langle \boldsymbol{w}_{\boldsymbol{accel}}^2\rangle+\langle \boldsymbol{w}_{\boldsymbol{shear}}^2\rangle}$<br>$\frac{\langle \boldsymbol{w}_{\boldsymbol{accel}}^2\rangle}{\langle \boldsymbol{u}^2\rangle} = 3(1-b)^2\frac{\gamma}{\gamma-1}\frac{(\theta_i+\theta_j)-4\theta_i\theta_j\sqrt{\frac{1+\theta_i+\theta_j}{(1+\theta_i)(1+\theta_j)}}}{(\theta_i+\theta_j)}\left(\frac{1}{(1+\theta_i)(1+\theta_j)}-\cdots\right.$<br>$\left.-\frac{1}{(1+\gamma\theta_i)(1+\gamma\theta_j)}\right)$, universal<br>$\frac{\langle \boldsymbol{w}_{\boldsymbol{shear}}^2\rangle}{\langle \boldsymbol{u}^2\rangle} = 0.238\left(\frac{\langle \boldsymbol{v}_{\boldsymbol{i}}^2\rangle}{\langle \boldsymbol{u}^2\rangle}\frac{\theta_i}{C_{c,i}}+\frac{\langle \boldsymbol{v}_{\boldsymbol{j}}^2\rangle}{\langle \boldsymbol{u}^2\rangle}\frac{\theta_j}{C_{c,j}}+\sqrt{\frac{\theta_i\theta_j}{C_{c,i}C_{c,j}}\frac{\langle \boldsymbol{v_i}\boldsymbol{v_j}\rangle}{\langle \boldsymbol{u}^2\rangle}}\right.$, universal$\left.\right)$<br>$\frac{\langle \boldsymbol{v}_{\boldsymbol{i}}^2\rangle}{\langle \boldsymbol{u}^2\rangle} = \frac{\gamma}{\gamma-1}\left(\frac{1+b^2\theta_i}{1+\theta_i}-\frac{1+b^2\gamma\theta_i}{\gamma(1+\gamma\theta_i)}\right), \quad \frac{\langle \boldsymbol{v}_{\boldsymbol{j}}^2\rangle}{\langle \boldsymbol{u}^2\rangle} = \frac{\gamma}{\gamma-1}\left(\frac{1+b^2\theta_j}{1+\theta_j}-\frac{1+b^2\gamma\theta_j}{\gamma(1+\gamma\theta_j)}\right)$, universal<br>$\frac{\langle \boldsymbol{v_i}\boldsymbol{v_j}\rangle}{\langle \boldsymbol{u}^2\rangle} = \frac{\gamma}{\gamma-1}\left(\frac{(\theta_i+\theta_j+2\theta_i\theta_j)+b(\theta_i^2+\theta_j^2-2\theta_i\theta_j)+b^2(\theta_i^2\theta_j+\theta_i\theta_j^2+2\theta_i\theta_j)}{(\theta_i+\theta_j)(1+\theta_i)(1+\theta_j)}-\cdots\right.$<br>$\left.-\frac{(\theta_i+\theta_j+2\gamma\theta_i\theta_j)+b\gamma(\theta_i^2+\theta_j^2-2\theta_i\theta_j)+b^2(\gamma^2\theta_i^2\theta_j+\gamma^2\theta_i\theta_j^2+2\gamma\theta_i\theta_j)}{\gamma(\theta_i+\theta_j)(1+\gamma\theta_i)(1+\gamma\theta_j)}\right),$<br>$\theta_i$ and $\theta_j \ll 1$<br>$\frac{\langle \boldsymbol{v_i}\boldsymbol{v_j}\rangle}{\langle \boldsymbol{u}^2\rangle} = \frac{2(1-b)^2}{\theta_i+\theta_j+\theta_j\theta_jW}-\frac{(1-b)^2}{\theta_i+\theta_j+\theta_j\theta_j(W+1)}+b(1-b)\left(\frac{1}{1+\theta_i+\frac{\theta_i}{2}W}+\frac{1}{1+\theta_j+\frac{\theta_j}{2}W}\right),$<br>$\theta_i$ or $\theta_j \gg 1$<br>$W = \sqrt{\frac{1+b^2\theta_i}{1+\theta_i}+\frac{1+b^2\theta_j}{1+\theta_j}}$ | (39) |

(continued on next page)

**Table 2** (*continued*)

| Reference | Mode | Frequency | |
|---|---|---|---|
| Wang et al. (1998) | Accelerative – correlated | $\beta = 2\sqrt{2\pi}(r_i + r_j)^2\left(\frac{1}{15}(r_i + r_j)^2\frac{\epsilon}{\nu} + \left(1 - \frac{\rho}{\rho_{i,j}}\right)^2\langle\left(\frac{Du_x}{Dt}\right)^2\rangle(\tau_i + \tau_j)^2 + \cdots\right.$<br>$\left. +2\left(1 - \frac{\rho}{\rho_{i,j}}\right)^2\tau_i\tau_j\langle\left(\frac{Du_x}{Dt}\right)^2\rangle\frac{(r_i+r_j)^2}{\lambda_D^2} + \frac{\pi}{8}(\tau_i + \tau_j)^2\left(1 - \frac{\rho}{\rho_{i,j}}\right)^2 g^2\right)^{1/2}$ | (40) |
| Zhou et al. (1998) | Accelerative – correlated | $\beta = 2\sqrt{2\pi}d_p^2\nu_p\left[1 - \exp\left(-\frac{1}{\theta}\right)\right]\sqrt{\frac{\theta}{1-\theta\left(1-\exp\left(-\frac{1}{\theta}\right)\right)}}$ | (41) |
| Mei and Hu (1999) | Orthokinetic | $\beta = \left\{1.2944^{2.2} + \left[\frac{1.3333\Gamma}{(\epsilon/\nu)^{1/2}}\right]^{2.2}\right\}^{1/2.2}(r_i + r_j)^3(\epsilon/\nu)^{1/2}$ | (42) |
| Reade and Collins (2000) | Preferential concentration | $\beta = 4\pi d^2 g(d)\int_{-\infty}^{0} -w_r P(w_r \mid d)dw_r$<br>$g(\hat{r}, \hat{d}, St) = 1 + c_0\hat{r}^{-c_1}e^{(-c_2\hat{r})} + \left(\frac{1}{3} + \frac{c_0 c_2^{c_1-3}}{\hat{d}^3}\int_0^{c_2\hat{d}} z^{2-c_1}e^{-z}dz\right) \times \cdots$<br>$\times\left((b_0 - b_1\sqrt{\bar{r} - 1})e^{-b_2(\bar{r}-1)} + \frac{b_3\delta}{\bar{r}^{3+\delta}}\right), \quad \bar{r} \geqslant 1$<br>$g(\hat{r}, \hat{d}, St) = 0, \quad \bar{r} < 1$<br>$b_0 = 56.7 \times \frac{St^{4/3}(1-0.957St)}{(1+19St^{10/3})}$<br>$b_1 = 80e^{-4St} \quad b_2 = 2St^{-5/3}$<br>$b_3 = 1 - \frac{b_0(2+2b_2+b_2^2)}{b_2^3} + \frac{\sqrt{\pi}b_1(15+12b_2+4b_2^2)}{8b_2^{7/2}}$<br>$c_0 = \frac{7.92St^{1.80}}{0.58+St^{3.29}} \quad c_1 = \frac{0.61St^{0.88}}{0.33+St^{2.38}} \quad c_2 = 0.25$ | (43) |
| Wang et al. (2000) | Preferential concentration | $\beta = 2\pi d^2 g(\ d)\langle\lvert w_r\rvert\rangle$<br>$\langle\lvert\ w_r\ \rvert\rangle = \left(\frac{2}{\pi}(\langle w_{r,accel}^2\rangle + \langle w_{r,shear}^2\rangle)\right)^{1/2}$<br>$\frac{\langle w_{r,accel}^2\rangle}{u'^2} = C_w\frac{2\gamma\theta}{\gamma-1}\left(1 - \frac{(1+2\theta)^{1/2}}{1+\theta}\right)\left(\frac{1}{(1+\theta)^2} - \frac{1}{(1+\gamma\theta)^2}\right)$<br>$\frac{\langle w_{r,shear}^2\rangle}{(\epsilon\nu)^{1/2}} = \frac{1}{15}\left(\frac{d}{\eta}\right)^2$<br>$\frac{g(d)-1}{Re_\lambda} = \frac{y_0(St)[1-z_0^2(St)]}{Re_\lambda} + z_0^2(St)\{y_1(St)[1 - z_1(St)] + y_2(St)z_1(St) + y_3(St)z_2(St)\}$<br>$y_0(St) = 18St^2, \quad St < 0.5$ | (44) |
| [10pt] | | $y_1(St) = 0.36St^{2.5}e^{-St^{2.5}}, \quad 0.5 < St < 1.25$<br>$y_2(St) = 0.24e^{-0.5St}, \quad 1.25 < St < 5$<br>$y_3(St) = 0.013e^{-0.07St}, \quad St > 10$<br>$z_0(St) = \frac{1}{2}\left[1 + \tanh\frac{St-0.5}{0.25}\right]$<br>$z_1(St) = \frac{1}{2}\left[1 + \tanh\frac{St-1.25}{0.1}\right]$<br>$z_2(St) = \frac{1}{2}\left[1 + \tanh\frac{St-6.5}{2.5}\right]$ | |
| Zhou et al. (2001) | Preferential concentration | $\beta = 2\pi\ (r_i + r_j)^2 g(r_i + r_j)\langle\lvert w_r\rvert\rangle$<br>$\langle\lvert w_r\rvert\rangle = \left(\frac{2}{\pi}(\langle w_{r,accel}^2\rangle + \langle w_{r,shear}^2\rangle)\right)^{1/2}$<br>$\langle w_{r,accel}^2\rangle = C_w(\alpha)\langle \boldsymbol{w}_{\boldsymbol{accel}}^2\rangle$<br>$C_w(\alpha) = 1.0 + 0.6e^{-(\alpha-1)^{1.5}}\alpha = \max[\theta_i/\theta_j; \theta_j/\theta_i]$<br>$\frac{\langle w_{r,shear}^2\rangle}{(\epsilon\nu)^{1/2}} = \frac{1}{15}\left(\frac{r_i+r_j}{\eta}\right)^2$<br>$g_{ij}(r_i + r_j) = 1 + \rho_{ij}^n[(g_{ii}(r_i + r_j) - 1)(g_{jj}(r_i + r_j) - 1)]^{1/2}$<br>$\rho_{ij}^n = y_1(St) + y_2(St)z(St)$<br>$y_1(St) = 2.6e^{-St} \quad 1 < St < 2.5$<br>$y_2(St) = 0.205e^{-0.0206St} \quad St > 2.5$<br>$z(St) = \frac{1}{2}[1 + \tanh(St - 3)]$ | (45) |
| Zaichik et al. (2006) | Preferential concentration | $\beta = 2\pi d^2\ g(d)\langle\lvert w_r\rvert\rangle$<br>$\langle\lvert w_r\rvert\rangle = \left(\frac{2}{\pi}S_{pll}\right)^{1/2}$ | (46) |
| Zaichik et al. (2006) | Preferential concentration | $\beta = 2\pi(r_i + r_j\ )^2 g_{i,j}(r_i + r_j)\langle\lvert w_r\rvert\rangle$<br>$\langle\lvert w_r\rvert\rangle = \left(\frac{2}{\pi}S_{pll}\right)^{1/2}$ | (47) |

3. The particles are of the same size or monodisperse.
4. Particle breakage is excluded.
5. Particles are spherical and remain so after collision.
6. Collisions only involve two particles (dilute suspension of particles).

Brownian motion is said to dominate the collision process for low particle Péclet numbers (<1, Benes et al., 2007), which contrasts the relative strengths of convection and diffusion. The particle Péclet number is expressed as

$$Pe = \frac{r\|\mathbf{V}\|}{D_o} = \frac{4\pi\left(\rho_{i,j} - \rho\right)gr^4}{3k_BT} \tag{3}$$

where $\|\mathbf{V}\|$ is the particle velocity magnitude, in this instance the Stokes velocity, where $D_o = k_BT/(6\pi\mu r)$ is the Stokes–Einstein coefficient of diffusion, $\rho_{i,j}$ the particle and $\rho$ the carrier fluid density, respectively and g the gravitational acceleration. It is assumed that the particle velocity vector, $\mathbf{V}$ is aligned in the gravitational direction.

### 2.2. Orthokinetic

Using the same assumptions as listed above, Smoluchowski (1917) developed Eq. (27) for laminar shear flows. The velocity gradient used in Eq. (27), $dU_x/dy$, is that for a flow exhibiting a simplified two-dimensional form of pure-shear strain where only one component of the relative velocity is considered.

Hu and Mei (1998) showed the result of Smoluchowski (1917) and an extensive list of other researchers to be inaccurate for monodisperse particles due to the inclusion of the self-collision.

However, for large particle numbers the distinction becomes negligible. One of the cited group of erring researchers, Collins and Sundaram (1998) demonstrates that their formulation (Sundaram and Collins, 1997) does indeed account for the self-counting effect.

In an attempt to generalize the work of Smoluchowski to three dimensions, Camp and Stein (1943) replaced the velocity gradient of Eq. (27) with a parameter $G$. For laminar flow, $G$ is a local parameter referred to as the absolute velocity gradient and is defined as

$$\begin{aligned}\Phi &= \mu G^2\\ &= \mu\left[\left(\frac{\partial U_x}{\partial y}+\frac{\partial U_y}{\partial x}\right)^2+\left(\frac{\partial U_x}{\partial z}+\frac{\partial U_z}{\partial x}\right)^2+\left(\frac{\partial U_y}{\partial z}+\frac{\partial U_z}{\partial y}\right)^2\right]\end{aligned} \tag{4}$$

where $\Phi$ is the viscous energy dissipation rate per unit volume.

For turbulent flow a global parameter, referred to as the root-mean-square velocity gradient, was defined and expressed as

$$G = \sqrt{\frac{\overline{\Phi}}{\mu}} \tag{5}$$

where $\overline{\Phi}$ is the mean value of the work input to the tank per unit time and unit volume. The overline indicates spatial averaging and thus $\overline{\Phi}$ is also referred to as the total spatial-averaged, steady, unit volume energy dissipation rate.

According to Pedocchi and Piedra-Cueva (2005) the validity of the Camp and Stein approach has been questioned. It has been shown to be locally inaccurate for general laminar flow as noted by Clark (1985), Saatçi and Halilsoy (1986), Kramer and Clark (1997) and Graber (1998). Pedocchi and Piedra-Cueva (2005) conclude that for laminar flows, Eq. (28), is valid, provided that the following expression for the viscous dissipation function is utilized

$$\begin{aligned}\Phi = \mu\Bigg[&2\left(\frac{\partial U_x}{\partial x}\right)^2+2\left(\frac{\partial U_y}{\partial y}\right)^2+2\left(\frac{\partial U_z}{\partial z}\right)^2+\left(\frac{\partial U_x}{\partial y}+\frac{\partial U_y}{\partial x}\right)^2\\ &+\left(\frac{\partial U_x}{\partial z}+\frac{\partial U_z}{\partial x}\right)^2+\left(\frac{\partial U_y}{\partial z}+\frac{\partial U_z}{\partial y}\right)^2\Bigg]\end{aligned} \tag{6}$$

Cleasby (1984) has shown Eq. (28) to be only valid for turbulent flows where the particle size is smaller than the Kolmogorov microscale, $\eta = (\nu^3/\epsilon)^{1/4}$. This stems primarily from the fact that $G$ includes the fluid viscosity, which only affects the viscous dissipation subrange associated with scales smaller than the microscale.

While Clark (1985) readily admits that, for a given steady turbulent flow field, a specific value of velocity gradient exists with which the correct average collision rate can be obtained using a Smoluchowski-type equation, it has never been shown that this gradient is simply related to the average energy dissipation rate. This is essentially what is claimed by Camp and Stein (1943).

Clark (1985) lists the following conditions that should apply to the use of Eq. (28)

1. Particles smaller than the Kolmogorov microscale.
2. Neutrally buoyant, spherical particles.
3. High Reynold's number, isotropic turbulence.
4. No large spatial variation in energy dissipation rate.
5. The coagulation process should be slow.
6. Normality in the distribution of $\partial u_x/\partial x$. Note that the local, instantaneous $x$-direction velocity component of a turbulent flow field, $U_x$, is expressed as $U_x = \langle U_x\rangle + u_x$ where the angled brackets, in this instance, indicate the time averaged velocity component whilst $u_x$ is the fluctuating, turbulent velocity component.
7. No hydrodynamic or colloidal interactions between particles ($\alpha = 1$).

A similar expression, Eq. (30), based on homogeneous isotropic turbulence with particles smaller than the Kolmogorov microscale, was derived by Saffman and Turner (1956) and also presented by Spielman (1978). This differs from Eq. (28) not only in the value of the constant used, 1.333 versus 1.294, but also in the definition of the rate of energy dissipation, where $\epsilon$ rather than $\overline{\Phi}/\rho$ is used.

According to Clark (1985) substitution of the instantaneous velocity components of a turbulent flow field, $U_x$, $U_y$ and $U_z$, into Eq. (6) and averaging over time yields the following expression for the total local energy dissipation, $E_t$

$$E_t = E_m + \epsilon \tag{7}$$

where $E_m$ is the dissipation associated with the mean flow ($\langle U_x\rangle$, $\langle U_y\rangle$ and $\langle U_z\rangle$) and $\epsilon$ that associated with the turbulent fluctuating components ($u_x$, $u_y$ and $u_z$).

Clark (1985) points out that for inhomogeneous turbulence, which is commonly associated with the flow field of flocculation devices, $E_m$ can be significant and Eq. (30) is thus expected to under estimate the collision rate.

Based on the results of their numerical investigation, where a spectral method was used to model the homogeneous turbulent flow field, Wang et al. (1998) note that Eq. (30) is only valid as long as the collided particles remain in the system and are allowed to overlap in space. It is also demonstrated that a non-overlapping particle formulation leads to a slightly higher collision rate (which increases with increased particle size) whilst the removal of collided particle pairs result in a lower value for smaller particle sizes than that predicted by Eq. (30).

Argaman and Kaufman (1968) developed a model for turbulent flocculation based on the hypothesis that the random motion of suspended particles can be characterized by a coefficient of diffusion. The latter was, in turn, expressed in terms of the energy spectrum of the turbulent velocity field. The form of the collision kernel is shown in Eq. (32) where $\langle u_x^2\rangle$ represents the mean-square velocity fluctuations and the angled brackets indicate the ensemble average. The energy spectrum coefficient, $K_s$ represents the effect of the shape of the turbulent energy spectrum on the coefficient of diffusion. The value of $K_s$ is determined primarily by the turbulent wave lengths or frequencies most pertinent to the flocculation process and should these frequencies be independent of the power input, $K_s$ will assume a constant value for a particular mixing device.

The mean-square velocity fluctuations are proportional to $G$. Experimental evidence seems to indicate that the constant of proportionality, $K_p$, or the paddle performance coefficient, is a constant for a particular mixing device. Note that $r_i$ represents the floc or aggregate size which is assumed to be significantly larger than the coagulating particles.

However, Cleasby (1984) notes that the use of $G$ in Eq. (32) was based on simplicity and familiarity rather than physical reasoning or even experimental verification as the latter was conducted at a constant temperature, i.e. viscosity remained constant. It is further demonstrated that, based on the experimental data, a good correlation between $\langle u_x^2\rangle$ and $(\overline{\Phi}/\rho)^{1/2}$ could as easily have been obtained.

Delichatsios and Probstein (1975) developed kinetic models for turbulent flocculation in a monodisperse system by applying simple binary collision mean free path concepts. Only isotropic turbulence is considered. The collision kernel is expressed by Eq. (34) where $w_x'$ or $\sqrt{\langle w_x^2\rangle}$ is the relative particle root-mean-square velocity that can be approximated by the root-mean-square relative turbulent velocity between two points at a distance of a particle diameter, $d$, apart. The $x$-direction is presumably aligned to the line that connects the colliding particles. The relative velocity depends on the turbulent scale where $\eta$ is the Kolmogorov microscale and $L$ the Eulerian macroscale of turbulence.

They tested their models experimentally but could unfortunately only verify their model for the case where $d < \eta$ due to simultaneous particle aggregation and breakup in the flow. It is worth noting that particle sizes comparable to the Eulerian macroscale of turbulence would necessarily imply neutrally buoyant particles in order to avoid inertial effects.

The following conclusions made by Cleasby (1984) warrant consideration

1. $G$ is only a valid parameter for flocculation of particles smaller than the microscale.
2. $(\overline{\Phi}/\rho)^{2/3}$ is a more appropriate flocculation parameter if particle sizes larger than the microscale are present.
3. In this range the agitator or mixing device might have an impact on flocculation as well as $(\overline{\Phi}/\rho)^{2/3}$, particularly at lower Reynold's numbers. Mixing devices should thus be evaluated individually to determine the best flocculation at any desired power input.
4. In this region of turbulent eddy flocculation, temperature should not play a role (fluid viscosity plays no significant role).

Based on their experimental work, Casson and Lawler (1990) state the likelihood that eddies responsible for flocculation will be about the same size as the particles being flocculated. The same sentiments were also put forward by Argaman and Kaufman (1968). Casson and Lawler (1990) stressed the fact that larger eddies contributed little to the flocculation process other than keeping particles in suspension. A method for estimating the velocity gradients in eddies of different sizes in the flow was developed and incorporated into Eq. (28), although the approach is thought to be valid only for oscillating flows.

Kramer and Clark (1997) demonstrated that for a discrete region of fluid exposed to linear velocity-gradients, the collision frequency of particles contained in the fluid region is a function of the strain rates acting on the volume element. A new scalar value, the absolute maximum principle strain rate, $|a'_{max}|$, is presented that accurately estimates the total collision rate. The collision kernel is shown in Eq. (38).

The model is used to numerically evaluate coagulation within a Couette flow apparatus. Results are contrasted to those obtained with the methodology of Camp and Stein, highlighting the deficiencies associated with their approach. A major obstacle in the use of Eq. (38) is that a detailed knowledge of the flow field is required before $|a'_{max}|$ can be calculated.

Using a numerical approach, Mei and Hu (1999) confirmed the results of Smoluchowski (1917) and Saffman and Turner (1956) for a uniform, laminar shear flow and Gaussian, isotropic turbulence, respectively. A formulation for the collision kernel is subsequently developed for rapidly sheared homogeneous turbulence resulting in Eq. (42), where $\Gamma$ is the constant shear rate imposed on the turbulent flow field. This formulation does away with the requirement of turbulence isotropy but does introduce an error of approximately 10% primarily due to the averaging of the transitional effect from isotropic to anisotropic turbulence.

### 2.3. Differential sedimentation

Under the influence of a gravitational field, particles of different sizes exhibit different terminal velocities which leads to collision and possible agglomeration.Eq. (29) was developed by Camp and Stein (1943) and reviewed by Lawler (1986). Although hydrodynamic and inter-particle forces have a significant effect on the collision characteristics during sedimentation, it is usually included through the collision efficiency.

Eq. (29) was also developed independently by Saffman and Turner (1956) and shown to be valid as long as the particles remain in the Stokes flow regime. According to the authors the particles should not be too dissimilar in size as hydrodynamic effects were not included in their analysis. Furthermore, the analysis focused on the coagulation of rain drops in clouds and as such assumes that the droplet or particle density far exceeds that of the carrier fluid as noted by Zhou et al. (1998).

As gravity is essentially an accelerative effect, it is most often associated with the analysis of the turbulent collision process where the inertia of the particles or drops plays a significant role, the work of Saffman and Turner (1956) being but one example.

### 2.4. Accelerative – correlated velocities

Saffman and Turner (1956) extended their analysis to include flows where particle inertia as well as gravity play a role, resulting in Eq. (31) where $u_x$ is the fluctuating part of the fluid velocity field surrounding the particles. Note that the $x$-direction is parallel to the line that connects the centers of the two colliding particles.

However, it should be noted that the inertial effects considered can at best be classified as moderate as the assumption of a collision efficiency of unity severly restricts the difference in particle sizes $(1/2 < r_i/r_j < 2)$. Furthermore, although the particle relaxation times differ (supposedly giving rise to their inertial collision response), they should be less than the Kolmogorov time scale, i.e. Stokes numbers less than unity.

According to Batchelor (1951) the velocity gradient term in Eq. (31) can be expressed as follows at high Reynolds numbers

$$\left\langle \left(\frac{Du_x}{Dt}\right)^2 \right\rangle = 1.3\left(\frac{\epsilon^3}{\nu}\right)^{1/2} \tag{8}$$

whilst the particle relaxation time for a particle of size $r_i$ is expressed as

$$\tau_i = \frac{2C_{c,i}(\rho_i - \rho)r_i^2}{9\mu} \tag{9}$$

where $C_{c,i}$, the Cunningham slip correction factor, assumes a value of unity in systems utilizing water as carrier fluid.

Saffman and Turner (1956) note that Eq. (31) does not reduce to Eqs. (29) and (30) in the case of zero turbulence and when only zero inertia particles are present, respectively. This discrepancy is attributed to a simplification introduced to ease the integration of the relative velocity probability function.

However, Wang et al. (1998) contend that the discrepancies should rather be attributed to the use of a cylindrical as opposed to a spherical formulation for the collision kernel. The underlying assumption being that the relative velocity at any instant is locally uniform over a spatial scale on the order of the collision radius, $(r_i + r_j)$, which is invalid for turbulent flows. It is also demonstrated that Eq. (31) overestimates the number of collisions by 25% in isotropic turbulence and by 20% for a simple uniform shear flow.

An improved version of the formulation of Saffman and Turner (1956) is proposed by Wang et al. (1998) and listed here as Eq. (40) where $\lambda_D$ is the longitudinal Taylor-type microscale of fluid acceleration as defined by Hu and Mei (1997).

Williams and Crane (1983) analysed the fluctuating relative motion of two solid particles or liquid drops in a turbulent gas flow where the particles are of intermediate size and exhibit velocities that are neither well-correlated nor completely independent. According to Kruis and Kusters (1997), the expressions for the variance of the relative fluctuating particle velocity, $\langle w_x^2\rangle$, presented by Williams and Crane (1983) are essentially one-dimensional and, assuming isotropy, is a factor 3 smaller than the variance of the three-dimensional, fluctuating relative velocity vector between two particles, $\langle \mathbf{w}^2 \rangle$.

The collision kernel and relevant velocity formulations are presented in Eq. (35). Note that only accelerative or inertial effects are included in this formulation with no provision being made for turbulent shear effects.

In Eq. (35) the dimensionless particle relaxation time, $\theta_i$, is defined as

$$\theta_i = \frac{\tau_i}{T_L} \tag{10}$$

where $T_L$ is the Lagrangian integral time scale. Whilst $\gamma$ is expressed as

$$\gamma = 2\left(\frac{L}{\lambda_g}\right)^2 \tag{11}$$

in which the longitudinal integral length scale, $L$, and the transverse Taylor microscale (lateral Lagrangian length scale), $\lambda_g$, are defined, respectively, as

$$L = u'_x T_L \tag{12}$$

$$\lambda_g = u'_x\left(\frac{15\nu}{\epsilon}\right)^{\frac{1}{2}} \tag{13}$$

with $u'_x$ the carrier fluid $x$-component rms velocity or standard deviation ($\sqrt{\langle u_x^2 \rangle}$).

The expressions developed by Williams and Crane (1983) are mainly for gaseous systems and do not include the added mass effect experienced by particles moving through a liquid environment such as water. Particle drag is based on Stokes flow and thus places a limit on the maximum particle size ($d$ < 100 μm).

Focusing on the viscous subrange of turbulence, Yuu (1984) derived an expression, Eq. (36), for the fluctuating relative velocity of two inertial particles. The analysis included the added mass effect experienced by particles in liquid systems as well as that of turbulent shear.

The added mass effect or so-called buoyancy effect is accounted for with the introduction of the coefficient, $b$, defined as

$$b = \frac{3\rho}{2\rho_{i,j} + \rho} \tag{14}$$

whilst $a_i$ and $a_j$ is the reciprocal of the particle relaxation times, $\tau_i$ and $\tau_j$, respectively.

According to Kruis and Kusters (1997) neither the formulation of Williams and Crane (1983) nor that of Yuu (1984) is applicable to liquid systems with particle sizes in the inertial subrange of turbulence. It is further noted that the universal solution of Williams and Crane (1983), listed as Eq. (35), does not reduce in the limit, to that developed for small particles ($\theta_i$ and $\theta_j \ll 1$). It also fails to reduce to the limit as calculated by Saffman and Turner (1956). The latter is a direct consequence of the definition of $\gamma$ used by Williams and Crane (1983).

Kruis and Kusters (1997) state that the formulation of Yuu (1984) would not be valid for very small particles as the exponential as opposed to the parabolic form of the Langrangian correlation was used which is not valid for the viscous subrange of turbulence. The accelerative term therefore does not reduce to the limit of small particles as derived by Saffman and Turner (1956).

Using the approach of Yuu (1984) for small particles and that of Williams and Crane (1983) for larger particles, Kruis and Kusters (1996, 1997) developed a universal expression for the collision kernel as shown in Eq. (39). The added mass effect is included in the formulation whilst the parabolic form of the Langrangian correlation is utilized, rendering the expression valid for both the inertial as well as the viscous subrange of turbulence. The formulation reduces to the limit of small particles as derived by Saffman and Turner (1956). The most notable assumptions are

1. The turbulence characteristics of the carrier fluid are not affected by the presence of particles. The formulation is thus valid where the particulate/fluid mass ratio is smaller than 0.1.
2. The Basset history term, which is only of significance where very small particles collide within a liquid medium, is neglected.
3. The turbulence is isotropic.
4. The particle drag is described by Stokes law which is normally not applicable in the large particle limit. In case of the latter, the formulation would provide an upper limit of the collision frequency.
5. For small particles the velocity gradient between particles is considered constant.
6. Differential sedimentation and Brownian motion is not included in the formulation.
7. Particles are larger than the mean free path of the fluid.

The turbulence constant, $\gamma$, is defined as

$$\gamma = 2\left(\frac{T_L}{\tau_L}\right)^2 \tag{15}$$

with $T_L$ the Lagrangian integral time scale defined as

$$T_L = 0.4\frac{L}{u'} \tag{16}$$

and $\tau_L$ the Langrangian time scale.

Eq. (15) can be expressed as a ratio of length scales following the example of Williams and Crane (1983) in Eq. (11) yielding

$$\gamma \cong \frac{L}{\lambda_f} \tag{17}$$

or

$$\gamma = 0.183\frac{u'^2}{\sqrt{\epsilon\nu}} \tag{18}$$

where $\lambda_f$ is the longitudinal Taylor microscale of fluid acceleration which, for isotropic turbulence, can be related to the transverse Taylor microscale in the following manner

$$\lambda_f = \sqrt{2}\lambda_g \tag{19}$$

It should be noted that the formulation of the relative particle velocity correlation, $\langle \boldsymbol{v_i v_j} \rangle$ , presented by Kruis and Kusters (1997) is not universal, in other words, there is no single expression that holds for both small and large particles. For example: in their 1996 publication, Kruis and Kusters (1996) make use of the small particle relative velocity correlation when demonstrating the use of their universal formulation for the particle collision frequency. Particle sizes range from 0.1 to 10 μm whilst the Kolmogorov length scale of the two turbulent flows under consideration is 150 μm and 1.67 mm, respectively.

Wang et al. (2000) contrast the normalized total relative velocity, $\langle |\mathbf{w}| \rangle / u'$, as a function of $\tau/T_e$, obtained through DNS with the expressions developed by Williams and Crane (1983) and Kruis and Kusters (1997) and report that the former under predicts the DNS values throughout the $\tau/T_e$-range whilst the latter is only accurate for $Re_\lambda$ = 24 and $\tau/T_e$ < 1. At higher $Re_\lambda$-values the normalized relative particle velocity correlation is under predicted.

Hu and Mei (1997) developed an expression, shown in Eq. (37), for the collision between moderately inertial particles along the lines of the Saffman and Turner (1956) analysis, although the effect of gravity was not included. A spherical as opposed to cylindrical formulation was utilized as well as a more appropriate relative particle velocity probability function (isotropic turbulence). Added mass effects were not included and as such the formulation is only valid for gaseous systems. The result of Saffman and Turner (1956) is recovered in the limit of small particles (orthokinetic collisions). An additional term is added to account for inertial collision between particles of the same size (monodisperse particles), the omission of which, according to Hu and Mei (1997), is a weakness of previous formulations.

Wang et al. (1998) extended the formulation of Hu and Mei (1997) to include gravity as well as a finite density-ratio correction, $(1-\rho/\ \rho_{i,j})$. The latter arises from the inclusion of the pressure-gradient force term in the equation of particle motion (e.g., Maxey and Riley (1983)). The result is listed in Eq. (40).

Wang et al. (1998) note that, apart from the corrections introduced by Hu and Mei (1997) Eq. (40) is superior to Eq. (31) in that the gravity term is formulated using the correct relative particle velocity probability function. One significant assumption is that no coupling of the gravitational effect with the other effects are considered.

Both Hu and Mei (1997) and Wang et al. (1998) acknowledge that their formulations do not include the influence of non-uniform particle concentration due to flow-particle microstructure interaction. From their numerical results, Hu and Mei (1997) conclude that inertia enhances the collision process through primarily two mechanisms, the first due to the increase in particle collision velocity and the second due to preferential particle concentration.

### 2.5. Preferential concentration

Preferential particle concentration first came to light during a numerical investigation of particle settling in cellular flow fields where Maxey and Corrsin (1986) noted that weakly inertial particles tend to collect along isolated paths. During further numerical investigation Maxey (1987) observed that inertial particles preferentially concentrate in regions of high strain rate and low vorticity, with the opposite being true for bubbles suspended in a more dense carrier fluid, Maxey (1987). The effect was also demonstrated through DNS, albeit at low Taylor Reynolds numbers, $Re_\lambda$, by Squires and Eaton (1990, 1991) and Wang and Maxey (1993) who showed that preferential concentration follows a Kolmogorov scaling, being most effective in producing a non-uniform concentration when $\tau_p/\tau_k \sim 1$.

Preferential concentration has been observed experimentally, most notably in a jet flow dominated by vortex ring structures, Longmire and Eaton (1992), in plane wake flows, Tang et al. (1992), in channel flows, Fessler et al. (1994) and in homogeneous and isotropic turbulence, Wood et al. (2005). Apart from Longmire and Eaton (1992) all of these investigations confirmed the Kolmogorov scaling introduced by Wang and Maxey (1993) as well as the concentration effect reaching a maximum at a Kolmogorov Stokes number in the vicinity of unity.

A number of studies have pointed to the multi-scale nature of the preferential concentration phenomenon at elevated $Re_\lambda$-values; Goto and Vassilicos (2006, 2008), Chen et al. (2006) and Yoshimoto and Goto (2007). DNS of preferential concentration is difficult at high $Re_\lambda$-values due to the prohibitive nature of the computational cost involved. Determining the scaling of the preferential concentration effect with $Re_\lambda$ is thus difficult.

Based on results obtained from DNS van Aartrijk and Clercx (2008), Collins and Keswani (2004) and Hogan and Cuzzi (1999, 2001) conclude that preferential concentration is only a weak function of $Re_\lambda$. The $Re_\lambda$-values considered in these investigations ranged from 40 to 200. Bec et al. (2007) argued that the weak $Re_\lambda$ dependence is only valid at dissipative scales but not within the inertial range where a much broader range of length and time scales are involved. These sentiments are echoed by Balkovsky et al. (2001) and Boffetta et al. (2004) with the latter concluding that to fully understand the geometry of preferential concentration, the presence of structures characterized by a large set of time scales cannot be ignored. Scott et al. (2009) introduced a clustering length scale, similar to the integral scale for fluid flow, which gives an indication of the spacing between particle clusters.

The effect of preferential particle concentration on the collision frequency can be dramatic with an increase of one to two orders of magnitude reported by Sundaram and Collins (1997). This result was obtained from an investigation making use of DNS. In fact, most investigations dealing with the formulation of collision kernels associated with the preferential particle concentration phenomenon are exclusively numerical. According to Wang et al. (2000) a numerical approach is preferable as it allows for the isolation and characterization of the different phenomena or factors influencing the collision process.

In their numerical investigation Sundaram and Collins (1997) considered the collision of heavy, monodisperse particles (all particles are identical) in a turbulent flow (isotropic) in the absence of global shear and gravity. The general collision kernel is formulated as

$$\beta = \frac{1}{2}\pi d^2 g(d) \int wP(\mathbf{w}|d)d\mathbf{w} \tag{20}$$

where $g(d)$ is the radial distribution function (RDF) and $P(\mathbf{w}|d)$ is the conditional relative velocity probability density function at contact. The former is a correction to the local number density function that accounts for the preferential concentration effect whilst the latter accounts for the decorrelation of adjacent particle motions.

Sundaram and Collins (1997) were able to demonstrate that the collision rate of finite inertia particles increased rapidly with increasing Stokes numbers, reaching a maximum at $St = 4$ and declining gradually thereafter. The collision rate is bounded in the lower limit by the expression of Saffman and Turner (1956) for orthokinetic collisions, Eq. (31) and that of Abrahamson (1975), Eq. (33) in the upper limit, a result confirmed by Chen et al. (1998), Chen et al. (1998). The formulation of Abrahamson (1975) is for particles with completely uncorrelated velocities and is discussed in the next section.

The initial rapid increase in the collision rate is shown to be the result of the combined effect of preferential concentration (reaching a maximum at $St = 0.4$) and the decorrelation of the velocities of adjacent particles. At elevated Stokes numbers the decorrelated particles struggle to obtain energy from the turbulent carrier fluid thus leading to a decline in the particle collision rate.

The results of Sundaram and Collins (1997) were confirmed in the numerical investigation conducted by Zhou et al. (1998) who also observed that the formulation of Abrahamson (1975) was only valid at very high $\tau_p/T_L$-values. Note that $T_L$ differs by a constant factor from the formulation used by Kruis and Kusters (1997)

$$T_L = \frac{v^2}{\epsilon} \tag{21}$$

An improvement of the formulation of Abrahamson (1975) for monodisperse heavy particles was developed, the so-called eddy-particle interaction (EPI) model, and is shown in Eq. (41) where $\theta$ is defined as

$$\theta = 0.5\frac{\tau_p}{T_L} \tag{22}$$

and $v_p$ is the rms particle fluctuating velocity.

Eq. (41) fits the numerical data accurately for $\theta$-values in excess of 1.5 and merges with the formulation of Abrahamson (1975) at elevated $\theta$-values. Note that the formulation is only valid for monodisperse particles in an isotropic turbulent flow in the absence of global shear and gravity. Furthermore, the formulation assumes that the particle density far exceeds that of the carrier fluid and that the particle diameter is less than the Kolmogorov microscale, $\eta$.

It is worth noting that the collision rate, $\dot{N}_c$, in a volume containing monodisperse particles is expressed as

$$\dot{N}_c = \beta \frac{N_p^2}{2} \tag{23}$$

where $N_p$ is the particle number density.

In their DNS investigation, Wang et al. (2000) investigate the effect of Stokes number and $Re_\lambda$ on the collision statistics of monodisperse particles. The effect of particle loading and particle diameter was not considered and particles nominally smaller than $\eta$ were analyzed. Although the particle concentration is well within the dilute limit, allowing a numerical treatment that leaves the fluid turbulence properties unaffected by the presence of the particles, preferential concentration could lead to local particle concentrations that would be high enough to influence the turbulence structure of the flow field. As such the collision kernel represents an upper limit of particle collision.

The collision kernel developed by Sundaram and Collins (1997), which is based on a cylindrical formulation, was corrected using the spherical approach which yielded the following expression for the collision kernel

$$\beta = 2\pi d^2 g(d)\langle w_r \rangle \tag{24}$$

where $w_r$ is the radial relative velocity between two colliding particles.

The numerical treatment allowed Wang et al. (2000) to isolate the effect of turbulent transport as quantified by $\langle |w_r| \rangle$, from the accumulation or preferential concentration effect as quantified by the radial distribution function at contact, $g(d)$.

Based on their numerical results an empirical model for the collision kernel was developed and is shown in Eq. (44) where $\theta = 2.5\tau_p/T_L$ and $\gamma = 0.183u'2/(\epsilon v)^{1/2}$. They obtained $C_w = 1.68$ by fitting the expression for $\langle |w_r| \rangle$ to the numerical data for $Re_\lambda = 58$. The radial distribution function was found to be a linear function of $Re_\lambda$, although the range considered was very limited, $Re_\lambda = 45$, 58 and 75.

Reade and Collins (2000) used DNS to determine the functional form and dependence of the RDF, $g(d)$. They were able to demonstrate that $g(d)$ could be decomposed as the product of two functions where the first depends on $Re_\lambda$ alone. As the effect of $Re_\lambda$ was not the primary concern of the investigation, the value of this function was set to unity and all simulations used for modeling purposes were performed at $Re_\lambda = 54.5$. It was further assumed that the particle density far exceeded that of the carrier fluid and that the system was dilute. The DNS formulation excluded any hydrodynamic effects (collision efficiency) and treated collisions as those between hard spheres (elastically rebounding). The functional dependence of the RDF was thus restricted to the particle diameter and Stokes number.

The functional form of the RDF is shown in Eq. (43) where $r = |\mathbf{x_1} - \mathbf{x_2}|$, $\hat{r} \equiv r/\eta$, $\hat{d} \equiv d/\eta$ and $\bar{r} \equiv r/d$. In the first expression for $g(\hat{r}, \hat{d}, St)$, the term containing $\delta$ was added to satisfy a specific integral constraint which is satisfied for all values of $\delta$. This term is negligible for small values of $\delta$. Note that the collision kernel is based on the spherical formulation of Wang et al. (1998) shown in Eq. (24) which relaxes the assumption of isotropy.

Zhou et al. (2001) extended their previous numerical investigation of the collision characteristics of monodisperse inertial particles, Wang et al. (2000), to that of bidisperse inertial particles. Of particular interest was the so-called accumulation effect (preferential concentration) as the particles from the two different size distributions could selectively respond to eddies of different sizes. The numerical experiments covered a $\tau_p/T_L$-value range of approximately 0–3 for both particle sizes with $Re_\lambda = 45$. In an effort to avoid small-scale features in the particle concentration fields at scales smaller than $\eta$, a $(r_i + r_j)/\eta$-value of unity was maintained. Collisions between particles from the two different size distributions alone were considered, yielding a $\beta^{DNS}$-value for each experiment. Zhou et al. (2001) plotted $\beta^{DNS}$-values, normalized by the collision kernel of Saffman and Turner (1956), for constant values of $\tau_i/T_L$ over the $\tau_j/T_L$-range and observed the following:

1. For small $\tau_i/T_L$-values the normalized collision kernel increases monotonically with $\tau_j/T_L$. At this scale, preferential concentration is negligible and the increase is due to an increase in the relative motion of particle size $j$.
2. For large $\tau_i/T_L$-values the normalized collision kernel decreases monotonically with $\tau_j/T_L$. Preferential concentration is again negligible and the decrease is due to the increased sluggish response to turbulent fluctuations of particle size $j$.
3. At intermediate $\tau_i/T_L$-values the normalized collision kernel increases with $\tau_j/T_L$, reaches a maximum and then decreases. This behavior is qualitatively consistent with a monodisperse system.
4. Where particle concentration is negligible, the bidisperse normalized collision kernel is larger than that of a monodisperse system.
5. Where particle concentration is important, the bidisperse normalized collision kernel is smaller than that of a monodisperse system, largely due to the lack of correlation between the particle concentration fields of the two particle sizes.
6. The RDF at contact of a bidisperse system, $g_{ij}$, attains a maximum when $\tau_i = \tau_j$ and is bounded from above by the smaller of the two monodisperse RDFs at contact, $g_{ii}$ and $g_{jj}$.
7. $\langle |w_r| \rangle$ in a bidisperse system is always larger than in a monodisperse system with the difference increasing as the difference in particle inertia increases.

Zhou et al. (2001) also compared normalized values of $\beta^{DNS}$ with values predicted by the collision models of Abrahamson (1975), Williams and Crane (1983), Kruis and Kusters (1997) and their eddy-particle interaction (EPI) model, Zhou et al. (1998). Note that none of these models make provision for particle concentration effects. The following is noted

1. The EPI model performs well if $\tau_i$ is of the order of $T_L$.
2. The model of Kruis and Kusters (1997) outperforms that of Williams and Crane (1983) although both under predict the normalized collision kernel.
3. The formulation of Abrahamson (1975) over predicts the collision kernel.
4. None of the models are accurate at scales where particle concentration is significant.

In developing their collision model, Eq. (45), the RDF at contact for the bidisperse system, $g_{ij}$, is related to that of two monodisperse systems, $g_{ii}$ and $g_{jj}$, through a correlation coefficient, $\rho_{ij}^n$. $g_{ii}$ and $g_{jj}$ can be calculated from the expressions in Eq. (44). A modified version of the relative velocity formulation of Kruis and Kusters (1997) where $\rho_p/\rho \gg 1$ is used to model $\langle w_{r,accel}^2 \rangle$ where $\alpha = \max[\theta_i/\theta_j;\ \theta_j/\theta_i]$. Note that $\theta = 2.5\tau_p/T_L$ and $\gamma = \alpha \times 0.183u'2/(\epsilon v)^{1/2}$.

Zhou et al. (2001) further notes that it can be assumed that $g_{ij}$ submits to the same linear Reynolds-number scaling as $g_{ii}$ and $g_{jj}$ and demonstrate that the model yields collision values within 20% of the numerical values for a rather narrow $Re_\lambda$ range of 45–58.

A method to model binary dispersion and accumulation of inertial particles in an isotropic turbulent flow field based on a kinetic equation for the probability density function (PDF) of the relative velocity of a pair of particles was developed by Alipchenkov and Zaichik (2003), Zaichik and Alipchenkov (2003) and Zaichik et al. (2006). This method is an extension of the one-point statistical approach to the two-point description and allows for the statistical modeling of distributions of the relative velocity of two particles in a random turbulent field as opposed to a stochastic description of particle motion along random trajectories. As a result the relative mean-square velocity, the radial distribution function and other two-particle statistical characteristics can be found.

The above approach yields an infinite set of balance equations which is closed at the second-order closure level invoking a gradient algebraic approximation for the triple fluctuating velocity correlations. A system of three non-linear ordinary differential equations with appropriate boundary conditions result. Solution of this set of equations yields the radial distribution function, $g(d)$ as well as the longitudinal and transverse components of the second-order particle velocity structure function, $S_{pll}$ and $S_{pnn}$, respectively.

The collision kernel formulated by Zaichik et al. (2006), shown here as Eq. (46), assumes that the particle density is much higher than that of the carrier fluid and also that the PDF of the relative velocity is Gaussian. Note that $\langle|w_r|\rangle$ is formulated in terms of $S_{pll}$ which results from the solution of the equation system.

Zaichik et al. (2006) solved the equation system and compared the results to those obtained from DNS investigations. A comparison of $\langle|w_r|\rangle$ over a range of $St$-values with the DNS results of Wang et al. (2000) showed good agreement. A comparison of the radial distribution function with the data of Sundaram and Collins (1997) and Wang et al. (2000) yielded the following:

1. The radial distribution function reduces to unity at very small and very large $St$-numbers as expected.
2. The radial distribution function goes through a peak as $St$ increases from very low values.
3. The peak corresponds to the Kolmogorov timescale.
4. The peak is slightly shifted towards particles with higher inertia.

The collision kernel of Eq. (46) demonstrates qualitative agreement with the DNS results of Wang et al. (2000) although the predicted maxima, much like the radial distribution function, are slightly shifted towards particles with higher inertia.

The model of Zaichik et al. (2006) was subsequently extended to include bidisperse inertial particles, Zaichik et al. (2006). As in the case of monodisperse particles, a system of three non-linear ordinary differential equations with appropriate boundary conditions are solved yielding the radial distribution function, $g_{i,j}(r_i + r_j)$ as well as the longitudinal and transverse components of the second-order particle velocity structure function, $S_{pll}$ and $S_{pnn}$, respectively. The collision kernel is shown in Eq. (47).

Zaichik et al. (2006) compared Eq. (47) with the DNS data of Zhou et al. (2001) and conclude that $\langle|w_r|\rangle$ compares well with the DNS results, especially at lower particle inertias. The slight discrepancy noted at higher inertias are thought to be the effect of particle inertia on the eddy-particle interaction time scales which is not accounted for in the collision kernel. It is also noted that the model responds well to changes in the Reynolds number, albeit over the limited range covered in the work of Zhou et al. (2001).

The radial distribution function at contact also compares favorably with the DNS data although the peak associated with preferential accumulation is slightly shifted to higher inertias. It is also noted that preferential accumulation is most pronounced in monodisperse systems, diminishing as particle inertias move further apart. As a result the collision rate in a monodisperse system may exceed that in a bidisperse system.

### 2.6. Accelerative – independent velocities

Under highly turbulent conditions particles from different turbulent eddies are projected into neighboring eddies leading to collision. According to Abrahamson (1975) classic kinetic theory can be used to develop a collision kernel under these circumstances.

A normal distribution of particle velocities is assumed. The particle velocity is expressed through the resolution of the Tchen equation where the Basset history term is omitted and an exponential form for the Lagrangian velocity correlation is assumed.

An expression for the collision frequency in the absence of any body forces was first developed after which Abrahamson (1975) postulated that, in the presence of body forces, an appropriate formulation can be developed by simply superimposing a constant terminal velocity on to the turbulent solution of the Tchen equation. The formulation for the collision frequency is shown in Eq. (33) where the $z$-direction corresponds to the direction of the resultant terminal velocities of the particle size ranges, $w_{ti}$ and $w_{tj}$ and $\langle u^2\rangle$, $\langle v_i^2\rangle$ and $\langle v_j^2\rangle$ are the one-dimensional, mean squared velocity deviation of the carrier fluid and the two particle sizes, respectively.

Note that the expression used to determine $\langle v_i^2\rangle$ and $\langle v_j^2\rangle$ in Eq. (33) follows from the analysis of Levins and Glastonbury (1972), with the additional assumption that the particle density is much larger than that of the carrier fluid which renders the formulation inappropriate for particulate-liquid systems.

Liepe and Möckel (1976) derived an expression for the particle and bubble velocity distribution variance in water from experimental work at intermediate Stokes numbers. This expression is commonly used, together with the formulation of Abrahamson (1975), to model flotation systems Schubert (1999), Bloom and Heindel (2002).

$$v_i' = \sqrt{\langle v_i^2\rangle} = 0.686\frac{\epsilon^{4/9} r_i^{7/9}}{\nu^{1/3}}\left(\frac{|\rho_i - \rho|}{\rho}\right)^{2/3} \tag{25}$$

The Tchen equation makes use of the Stokes drag law and as such severely restricts the upper limit of the particle size range. For higher particle Reynolds numbers Eq. (33) will thus overestimate the number of collisions.

Abrahamson (1975) also developed an expression for the lower limit of the particle size that would ensure independent particle velocities. The expression for the $i$th particle size is

$$r_i^2 = \frac{15\nu\langle v_i^2\rangle}{4\rho\epsilon} \tag{48}$$

## 3. Summary

The development of particle collision models spans a period of close to a 100 years and includes models that range from simplistic, chosen for their mathematical convenience rather than accuracy, to highly involved models that attempt to include complex particle-fluid interactions such as preferential concentration.

The experimental analysis of collision processes is difficult and becomes even more so as the complexity of the carrier fluid flow field increases. As a result, collision models for relatively simple flow conditions associated with perikinetic collisions (Smoluchowski, 1917), differential sedimentation and orthokinetic collisions for simple laminar shear flow ( Camp and Stein, 1943) were formulated relatively early on in the development process and remain largely unchallenged to this day.

However, a vast number of industrial processes where particle collision is of importance is characterized by turbulent flow. The boundaries of this collision regime is fairly well demarcated on the one side with the formulation of Saffman and Turner (1956) and on the other by that of Abrahamson (1975). The former applies to particles that are significantly smaller than the smallest scale of turbulence and exhibit velocities which are perfectly correlated with that of the surrounding carrier fluid whilst the latter considers particles with very high inertias, exhibiting velocities that are completely decorrelated from that of the surrounding carrier fluid.

Considerable effort has been expended in the development of a so-called universal collision model that would be valid over the entire range of particle inertias. Arguably the most well-known would include the formulations of Williams and Crane (1983) and Kruis and Kusters (1997). However, the consensus is that the model of Williams and Crane (1983) and that of a number of other authors are erroneously based on a cylindrical as opposed to a spherical formulation and do not recover the results of Saffman and Turner (1956) in the limit of small particles. Furthermore, Hu and Mei (1997) note that formulations of all previous authors ignore collisions between identical particles. The formulation of Wang et al. (1998) not only remedies the above-mentioned short-comings but extends the formulation to include non-gaseous carrier fluids.

The preferential concentration of particles that exhibit relaxation times close to the Kolmogorov time scale, in regions of high strain rate and low vorticity has a marked effect on particle collision. Reported increases of the collision frequency range from one to two orders of magnitude.

Attempts at formulating a collision kernel that includes the effect of preferential concentration are primarily based on the formulation proposed by Sundaram and Collins (1997) where the radial distribution function at contact is used to account for the concentration effect and the relative velocity probability density function for the decorrelation of adjacent particle motion.

The cylindrical formulation of Sundaram and Collins (1997), now considered erroneous, was corrected by Wang et al. (2000), introducing the spherical formulation of the collision kernel. Zhou et al. (2001) extended the work of previous authors on preferential concentration and most notably that of Wang et al. (2000) to include bidisperse as opposed to monodisperse particles.

It is worth noting that the above-mentioned collision models that include the effect of preferential concentration are all based on collision data obtained through DNS rather than experimental data. Numerical limitations place a severe restriction on the magnitude of the turbulent flow Reynolds number that can be modeled and as such the effect thereof on the collision process remains elusive.

Moving away from DNS, Alipchenkov and Zaichik (2003), Zaichik and Alipchenkov (2003) and Zaichik et al. (2006) used the resolution of the kinetic equation for the probability density function of the relative velocity of a pair of particles to formulate the collision kernel for both monodisperse as well as bidisperse particles. Good agreement is obtained with the DNS data of other authors.

In conclusion it is well worth noting that the development of collision models have come to rely near exclusively on DNS rather than experimental data sets. The numerical modeling of the collision process holds several advantages of which the ability to control the primary variables as well as the accuracy with which collisions can be monitored are foremost. However, the move towards DNS also imposes severe restrictions as noted above which would suggest that the pace of further development of collision models will be governed by the increase in computational resources.

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