Exponential Scaling in Early-stage Agglomeration of Adhesive Particles in Turbulence

Sheng Chen,1 Shuiqing Li,1,* and Jeffrey S. Marshall2

1Department of Energy and Power Engineering, Tsinghua University, Beijing 100084, China.
2Department of Mechanical Engineering, The University of Vermont, Burlington, VT 05405, USA.
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We carry out direct numerical simulation together with an adhesive discrete element method calculation (DNS-DEM) to investigate agglomeration of particles in homogeneous isotropic turbulence (HIT). We report an exponential-form scaling for the size distribution of early-stage agglomerates, which is valid across a wide range of particle inertia and inter-particle adhesion values. Such scaling allows one to quantify the state of agglomeration using a single scale parameter. An agglomeration kernel is then constructed containing the information of agglomerate structures and adhesion-controlled sticking probability. The kernel function extends Smoluchowski’s theory to the condition of non-coalescing solid adhesive particles and can reproduce DNS-DEM results with a simple one-dimensional model.

* Corresponding author: lishuiqing@tsinghua.edu.cn
I. INTRODUCTION

Clustering of particles suspended in turbulence has been extensively studied in experiments [1, 2], in simulations [3] and by theoretical approaches [4, 5]. To predict the evolution of cluster or agglomerator size, Smoluchowski’s equation, built on statistical collision kernels, is one of the few theoretical tools that can be applied to large-scale systems [6-8]. For particles in turbulence, the collision kernel is usually expressed as the production of the mean relative radial velocity and the radial distribution functions (RDFs) of particle pairs at the distance of contact. For zero-inertial particles, these two quantities can be statistically determined from those of turbulence flows [9]. In contrast, inertial particles preferentially sample certain regions of the flow due to the centrifugation effect, giving rise to higher values of both relative radial velocity and spatial concentration [10-14]. As the inertia of particles further increases, particles from different regions of the flow come together. A larger relative velocity, consequently a larger collision rate, is then observed. Such effect is termed as ‘caustics’ [15] or ‘sling effect’ [16].

Based on these models of geometric collision kernel, Smoluchowski’s theory can be then used to describe the growth of clusters assuming that colliding particles merge immediately to form new larger spherical particles. The assumption of unity coagulation efficiency is normally valid for droplets. However, it is not applicable to the agglomeration of solid non-coalescing adhesive particles. Such systems are quite ubiquitous, ranging from electrostatic agglomerators [18], flocculation during water treatment [19], assemblage of preplanetary grains [20] to the growth of dendrites during aerosol filtration [21, 22]. The solid adhesive particles, across 1 to 10$^5$ microns, have two significant differences from Brownian nanoparticles or coalescing droplets: (1) the interparticle adhesion due to van der Waals attraction is short-ranged and relatively soft [23]. It leads to the sticking/rebound behavior of colliding particles (i.e., non-unity coagulation efficiency). (2) Formed agglomerates are usually non-spherical, whose structure will evolve due to restructuring and breakage. It has been reported that even the simplest elastic repulsion between particles considerably changes the picture of agglomeration [24]. Constructing a kernel function that can reflect the influence of complicated inter-particle interactions is a crucial problem that has not been settled.

Solving this problem requires a fundamentally different approach - discrete element methods (DEM) - that tracks the dynamics of individual particles both while they are traveling alone through the fluid and while they are colliding with other particles [23]. To properly simulate the agglomeration, particle collisions should be resolved with a time step much smaller than the Kolmogorov time scale. Moreover, all the possible modes of particle interaction, i.e., normal impact, sliding, twisting, and rolling, should be taken into account [25, 26]. Constructing kernel functions or stochastic agglomeration models [27, 28] based on data from DEM simulations then allows large-scale simulation of the agglomeration process.

In this work, we perform direct numerical simulations (DNS) to study the agglomeration of non-coalescing solid particles in homogeneous isotropic turbulence (HIT) with focus on the effect of van der Waals adhesion. A novel adhesive DEM is employed to fully resolve the translational and rotational motions of particles. We report an exponential-form scaling for the size distribution of early-stage agglomerates as $n(A)/n_0 \sim \exp(-A/\kappa)$, where $n(A)$ is the number density of agglomerates of size $A$. This exponential distribution allows one to describe the growth of agglomerates using a single scale parameter $\kappa$. Based on the simulation results, we are able to extend the Smoluchowski’s theory to describe adhesion-enhanced agglomeration by introducing a turbulence agglomeration kernel depending on the fractal structure of agglomerates and an adhesion-controlled sticking probability.

II. NUMERICAL METHOD AND SIMULATION CONDITIONS

A. DNS-DEM

1. Fluid phase

In our simulation, the homogeneous isotropic turbulent flow is calculated by DNS on a cubic, triply-periodic domain. A pseudospectral method with second-order Adams-Bashforth time stepping is applied to solve the continuity and momentum equations of the incompressible flow,

$$\nabla \cdot \mathbf{u} = 0,$$

$$\frac{\partial \mathbf{u}}{\partial t} = \mathbf{u} \times \mathbf{\omega} - \nabla \left( \frac{p}{\rho_f} + \frac{\mathbf{u}^2}{2} \right) + \nu \nabla^2 \mathbf{u} + \mathbf{f}_F + \mathbf{f}_P. \tag{1b}$$

Here, $\mathbf{u}$ and $\mathbf{\omega}$ are the fluid velocity and vorticity, respectively. $p$ is the pressure, $\rho_f$ is the fluid density, $\nu$ is the kinematic viscosity. The small wavenumber forcing term $\mathbf{f}_F$ is used to maintain the turbulence with a approximately constant kinetic energy. As suggest in [29, 30], we assume the forcing vector to be propotional to the fluid velocity.
and added to wavenumbers with magnitude \( k < 5 \). \( f_p \) is the particle body force, which is calculated at each Cartesian grid node \( i \) using \( f_p(x_i) = -\sum_{p=1}^{N} F_{n}^{p} \delta_{h} (x_i - X_{p,n}) \). Here, \( x_i \) is the location of grid node \( i \), \( F_{n}^{p} \) is the fluid force on particle \( n \) located at \( X_{p,n} \) and \( \delta_{h} (x_i - X_{p,n}) \) is a regularized delta function. The two-way coupling between particles and fluid is adopted to reflect the effect of flow disturbance on agglomeration, which is non-negligible even when the particle volume fraction \( \phi < 0.01 \) [26].

Before the particles are added into the domain, a preliminary computation is conducted for 5000 time steps with \( dt = 0.005 \) to allow the turbulence to reach the statistically stationary state. The turbulence kinetic energy \( q \) and dissipation rate \( \epsilon \) are obtained from integration of the power spectrum \( E(k) \),

\[
q = \int_{0}^{k_{\text{max}}} E(k) \, dk, \quad \epsilon = 2\nu \int_{0}^{k_{\text{max}}} k^2 E(k) \, dk.
\]  

(2)

2. Solid phase: adhesive discrete element method

We use discrete element method (DEM) to model the particles’ motion in turbulent flows, which solves the linear and angular momentum equations of particles

\[
m_i \ddot{v}_i = F_i^F + F_i^C, \quad I_i \ddot{\Omega}_i = M_i^F + M_i^C,
\]  

(3a)

and

\[
F_i^F = -3\pi \mu d_p^2 (v - u) \, f, \quad M_i^F = -\pi \mu d_p^3 \left( \Omega - \frac{1}{2} \omega \right),
\]  

(4a)

where \( m_i \) and \( I_i \) are mass and moment of inertia of particle \( i \) and \( v_i \) and \( \Omega_i \) are the translational velocity and the rotation rate of the particle. The forces and torques are induced by both the fluid flow (\( F_i^F \) and \( M_i^F \)) and the interparticle contact (\( F_i^C \) and \( M_i^C \)). In this work, the dominant fluid force is the Stokes drag given by

\[
f = (1 - \phi)^{1-\zeta} \gamma = 3.7 - 0.65 \exp \left[ -\frac{1}{2} (1.5 - \ln Re_p)^2 \right].
\]

(5)

In addition to the Stokes drag, we also include the Saffman and Magnus lift forces in \( F_i^F \) [32][33].

When two particles \( i \) and \( j \) are in contact, the normal force \( F^N \), the sliding friction \( F^S \), the twisting torque \( M^T \), and the rolling torque \( M^R \) acting on particle \( i \) from particle \( j \) can be expressed as

\[
F_{ij}^N = F_{ij}^{NE} + F_{ij}^{ND} = -4F_C \left( \frac{a_{ij}^3}{3} - \frac{a_{ij}^{3/2}}{2} \right) \eta_N v_{ij} \cdot n_{ij},
\]

(6a)

\[
F_{ij}^S = -\min \left[ k_T \int_{t_0}^{t} v_{ij}(\tau) \cdot \xi_S \, d\tau + \eta_T v_{ij} \cdot \xi_S, \, F_{ij,\text{crit}}^S \right],
\]

(6b)

\[
M_{ij}^T = -\min \left[ k_T \int_{t_0}^{t} \Omega_{ij}(\tau) \cdot n_{ij} \, d\tau + \frac{\eta_T a_{ij}^2}{2} \Omega_{ij}^T \cdot n_{ij}, \, M_{ij,\text{crit}}^T \right],
\]

(6c)

\[
M_{ij}^R = -\min \left[ k_C \frac{a_{ij}^{3/2}}{3} \int_{t_0}^{t} \xi_L \cdot \Omega_{ij}^T \cdot n_{ij} + \eta_R v_{ij} \cdot t_R, \, M_{ij,\text{crit}}^R \right].
\]

(6d)

The normal force \( F_{ij}^N \) contains an elastic term \( F_{ij}^{NE} \) derived from the JKR (Johnson-Kendall-Roberts) contact theory and a damping term \( F_{ij}^{ND} \), which is proportional to the rate of deformation. \( F^ne \) combines the effects of van der Waals attraction and the elastic deformation and its scale is set by the critical pull-off force, \( F_C = 3\pi R_{ij} \gamma \), where \( R_{ij} = (r_{p1}^{-1} + r_{p2}^{-1})^{-1} \) is the reduced particle radius and \( \gamma \) is the surface energy of the particle. \( a_{ij} \) is calculated inversly from the normal particle overlap, \( \delta \), through

\[
\delta = 6 \frac{\delta_C}{a_{ij}} = 6 \left[ 2(\bar{a}_{ij})^2 - \frac{4}{3}(\bar{a}_{ij})^2 \right],
\]

(7)
where $\delta_C$ is the critical overlap. For details, see [25]. The sliding friction $F^S_{ij,crit}$, twisting torque $M^T_{ij,crit}$, and rolling torque $M^R_{ij,crit}$ (Eq. (6b) - (6d)) are all calculated using spring-dashpot-slider models, where $v_{ij}$, $\xi_{ij}$, $\Omega_{ij}$, and $\tau_{ij}$ are the relative sliding, twisting, and rolling velocities. When these resistances reach their critical limits, $F^S_{ij,crit}$, $M^T_{ij,crit}$ or $M^R_{ij,crit}$, a particle will irreversibly slide, twist or roll relative to its neighboring particle. The critical limits are expressed as [25]:

$$F^S_{ij,crit} = \mu F_C \left| 4 \left( \dot{\alpha}_{ij} - \dot{\alpha}_{ij}^{3/2} \right) + 2 \right|,$$  

$$M^T_{ij,crit} = \frac{3\pi a_{ij} F^S_{ij,crit}}{16},$$  

$$M^R_{ij,crit} = 4F_C \dot{\alpha}_{ij}^{3/2} \theta_{crit} R_{ij}.$$  

Here $\mu(=0.3)$ is the friction coefficient and $\theta_{crit}(=0.01)$ is the critical rolling angle. We set these values according to experimental measurements [34].

It is known that the fluid squeeze-film between particles near contact significantly reduces the approach velocity and further influences the collision and agglomeration process. In this work, viscous damping force derived from the classical lubrication theory is also included, given by

$$F_I = -\frac{3\pi \mu r_p^2}{2h} \frac{dh}{dt}.$$  

$F_I$ is initiated at surface separation distance $h=0.01r_p$ and a minimum value $h=2 \times 10^{-4}r_p$ is set at the instant of particle contact according to experiments [35][36].

3. Multiple-time step framework

Our DNS-DEM computational framework is designed with multiple-time steps [23][25][37]. The flow field is updated using a fluid time step $dt_F = 0.005$. To correctly identify inter-particle collisions, a smaller particle convective time step $dt_p = 2.5 \times 10^{-4}$ is adopted to update the force, velocity, and position of particles that do not collide with other particles. Such a small $dt_p$ ensures that the distance each particle travels during a time step is only a small fraction of the particle or the grid size. In addition, we build a local list at each fluid step to record the neighboring particles that each particle may collide as it is advected over a fluid time step. Once a particle is found to collide with other particles during a particle time step, we then recover its information (i.e., its force, velocity, and position) to the start of this particle time step and instead advect it using a collision time step $dt_C = 6.25 \times 10^{-6}$. The value of $dt_C$ is small enough to resolve the rapid variation of contacting forces, velocity, and position of the particles.

B. Simulation conditions

The system studied in this work is illustrated in Fig. 1. We consider $N = 4 \times 10^4$ non-Brownian solid particles suspended in the homogeneous isotropic turbulent flow. The triply periodic computational domain has a dimension of $(2\pi)^3$ with $128^3$ grid points. The Taylor Reynolds number is fixed as $Re_\lambda = 93.0$ in this work. Similar values of $Re_\lambda$ have also been used in previous studies involving particle-laden flows [12][38][39]. By setting this value of $Re_\lambda$, we can easily compare our results with those in literature. Other dimensionless flow parameters, including the fluctuating velocity $u'$, the dissipation rate $\epsilon$, the kinematic viscosity $\nu$, Kolmogorov length $\eta$, Kolmogorov time $\tau_k$, and the large-eddy turnover time $T_e$, are listed in Table I.

The particle radius is fixed as $r_p = 0.01$. We choose the value of particle radius so that the particle size and the Kolmogorov length scale are comparable. We choose this relatively large value of particle size to increase the collision rates, ensuring good statistics on agglomeration within a feasible computing time. The particle volume concentration $\phi = \frac{4N\pi r_p^3}{3}$ is small so that the system can be regarded as a dilute system. The particle density $\rho_p$, determined by the particle response time, satisfies $\rho_p \gg \rho_f$, with $\rho_f$ being the fluid density.

One of the most important parameters governing the agglomeration is the Kolmogorov-scale Stokes number, $St_k = \tau_p/\tau_k$, where $\tau_p = m_p/(6\pi r_p \mu)$ is the particle response time and $\tau_k = (\nu/\epsilon)^{1/2}$ is the Kolmogorov time. In the classical theory of turbulent collision of nonadhesive particles, $St_k$ significantly influences the value of the collision kernel. In the presence of adhesion, the adhesion parameter $Ad = \gamma/(\rho_p u'^2 r_p)$, defined as the ratio of interparticle adhesion to particle inertia, is normally used to quantify the adhesion effect [25]. The particle surface energy $\gamma$ is determined according to experimental measurements [34][10] or calculated from the Hamaker coefficients of the materials [25].
FIG. 1. Snapshot of the simulated system at \( t = 20 \). The enlarged view from the middle slice \((x = 0)\) shows agglomerates with size \( A \) (indicated by the colorcode) up to 8 particles.

TABLE I. Dimensionless parameters of the fluid turbulence, including the fluctuating velocity \( u' \), the dissipation rate \( \epsilon \), the kinematic viscosity \( \nu \), Taylor-mircoscale Reynolds number \( Re_\lambda \), Kolmogorov length \( \eta \), Kolmogorov time \( \tau_k \), and the large-eddy turnover time \( T_e \).

| \( u' \)  | \( \epsilon \)  | \( \nu \)  | \( Re_\lambda \) | \( \eta \)  | \( \tau_k \) | \( T_e \) |
|---------|-------------|---------|----------------|---------|---------|--------|
| 0.28    | 0.0105      | 0.001   | 93.0           | 0.0175  | 0.31    | 7.4    |

\( Ad \) has been successfully used to estimate the critical sticking velocity of two colliding particles \([41]\) and predict the packing structure of adhesive particles \([22, 42, 43]\). In this work, we systematically vary \( Ad \) to show the effect of adhesion on agglomeration.

C. Smoluchowski’s theory

Before showing the DNS-DEM results, we introduce the Smoluchowski coagulation equation and discuss how to apply the theory to the agglomeration of non-coalescing adhesive particles. In Smoluchowski’s theory, the growth of agglomerates can be described using the population balance equation (PBE)

\[
\dot{n}(A) = \frac{1}{2} \sum_{i+j=A} \Gamma(i, j) n(i) n(j) - n(A) \sum_{i=1}^{\infty} \Gamma(i, A) n(i),
\]

where \( \Gamma(i, j) \) is the rate constants (kernel) for an agglomerate of size \( i \) colliding with agglomerate of size \( j \) and should reflect all the factors affecting agglomeration. It is defined as \( \Gamma(i, j) \equiv \dot{n}_{c,ij}/(n(i)n(j)) \) with \( \dot{n}_{c,ij} \) being the collision rate per unit volume and \( n(i) \) being the average number concentration of size group \( i \). The first term in the right-hand side of Eq. (10) is the source term that accounts for the rate at which agglomerates of size \( A \) are created. The second term is a sink that describes agglomerate disappearance due to its coalescence with other agglomerates.

PBE can be readily used to predict the growth of droplets in clouds with an underlying assumption - colliding particles coalesce instantaneously to form larger particles \([44]\). Therefore, the growth rate of agglomerates is equivalent to the collision rate. The collision between adhesive non-coalescing microparticles, however, does not ensure the growth of an agglomerate. Both sticking and rebound could happen as a result of the competition between the particles’ kinetic energy and the surface energy. Thus, it is natural to introduce a sticking probability, \( \Theta \), defined as the ratio of the number of collisions that lead to agglomeration to the total number of collisions. We then have an agglomeration kernel, which reads

\[
\Gamma_a(i, j) = \Theta \Gamma(i, j), \quad \forall i, j.
\]

The sticking probability has a minimum value 0 for non-adhesive particle systems and a maximum value 1, corresponding to the hit-and-stick case in conventional PBE simulations. We can then simulate the agglomeration with
different adhesion level, by simply replacing $\Gamma(i,j)$ in Eq. (10) by $G_a(i,j)$. We will show below that such simple modification can well reproduce DNS-DEM results in a statistical manner.

The structure of agglomerates is another crucial factor affecting the agglomeration rate. For non-coalescing adhesive particles, the formed agglomerates usually have fractal structures, which distinguishes our system from those of droplets [26, 45]. In systems involving Brownian nanoparticles, theoretical collision kernels can be extended to fractal agglomerates when substituting the particle radius with the radius of effective collision spheres (ECSs) for an agglomerate [46, 47]. We will show below that the idea of the effective radius can also be applied to non-Brownian inertial particles.

III. RESULTS AND DISCUSSIONS

A. Collision rate, agglomerate size and structure

We first measure the temporal evolution of the collision kernel in a system with $St_k = 5.8$ and $Ad$ varying from 0.013 to 128. To show the adhesion effect, here we simply regard the system as a monodisperse system and count the collisions between every primary particle. The collision kernel is then calculated as $\Gamma = \frac{2n_c}{n_0^2}$, where $n_0$ is the number density of primary particles. The temporal evolution of the collision kernel $\Gamma(t)$, normalized by the collision kernel for zero-inertia particles $\Gamma_0 = (8\pi\epsilon/15\nu)^{1/2}(2r_p)^3$ [49], is shown in Fig. 2(a). When the adhesion is extremely weak ($Ad = 0.013$ and 1.3), the collision kernel rapidly reaches a statistically steady state with $\Gamma(t)/\Gamma_0 = 11.1$, which is consistent with previous DNS results for nonadhesive particles [12]. As $Ad$ increases, the collision kernel is significantly reduced and the system is pushed away from equilibrium. Since adhesion number only affects the interaction between contacting particles, we attribute these phenomena to adhesion-enhanced agglomeration. In Fig. 2(b), the agglomeration at $t = 15$ is clearly displayed in the form of the fraction of particles $P(A)$ contained in an agglomerate of size $A$ (number of particles in the agglomerate). For small $Ad$, most particles remain as singlets and only a small portion of particles ($\sim 4\%$) are contained in agglomerates of size $A \geq 2$. In contrast, a strong adhesion remarkably enhances the agglomeration, yielding a considerable number of agglomerates with size $A$ up to 20.

To model the agglomeration process in the framework of Smoluchowski’s equation, a measure of agglomerate structure in the form of the equivalent sphere is necessary. One such quantity is the radius of gyration, defined for an agglomerate of size $A$ by $R_g(A) = (\sum_i^A |\mathbf{X}_i - \mathbf{X}_g|^2/A)^{1/2}$, where $\mathbf{X}_i$ denotes the position of $i$th particle within the agglomerate and $\mathbf{X}_g$ is the centre of mass of the agglomerate. In Fig. 2(d), we show an agglomerate generated from DNS-DEM simulation and its equivalent sphere with the radius of gyration. We calculate $R_g$ for all the agglomerates produced in the simulations in Fig. 2(a) at $t = 15$ and plot the ratio $R_g/r_p$ as a function of agglomerate size $A$ (large size agglomerates with $A > 12$ only contain $0.2\%$ particles thus are neglected here). The results fall onto a power-law curve

$$ \frac{R_g(A)}{r_p} = \left( \frac{A}{k} \right)^{D_f} $$

with the factor $k = 1.75$ and the fractal dimension $D_f = 1.57$. The $D_f$ value measured here is within the range given by experimental measurements [48]. In this work, we focus on the agglomeration at early-stage, when the restructuring and breakage of agglomerates are normally not involved [48]. These phenomena will lead to a variation of factor $k$ and the fractal dimension $D_f$ [49], which is left for future work.

B. Effect of Stokes number

The temporal evolution of the collision kernel $\Gamma(t)/\Gamma_0$, the fraction of particles, $P(A)$, contained in agglomerates of size $A$, and the gyration radius of agglomerates $R_g(A)/r_p$ for cases with different Stokes number $St_k$ and adhesion parameter $Ad$ are plotted in Fig. 3. For particles with small inertia $St_k = 0.72$, the increase of adhesion parameter only has a limited effect on the temporal evolution of the collision kernel (Fig. 3(a)). The reason is that the system has a small value of collision rate at this $St_k$ and only a small portion of particles can form agglomerates upon collisions. The system still behaves as a monodisperse system. This is further displayed in the form of the fraction of particles $P(A)$ contained in an agglomerate of size $A$ (Fig. 3(b)). For particles with higher Stokes number $St_k = 5.8$ in Fig 2. For all the three $St_k$, the radius of gyration for agglomerates of different size can be well described using the power-law function in Eq. (12), with $k$ and $D_f$ insensitive to the value of adhesion parameter $Ad$. 

FIG. 2. (a) Temporal evolution of the collision kernel $\Gamma(t)/\Gamma_0$ for cases with $St_k = 5.8$ and $Ad = 0.013$ (circles), 1.3 (left-pointing triangles), 13 (diamonds), 64 (upward triangles), and 128 (squares). (b) Fraction of particles, $P(A)$, contained in agglomerates of size $A$ at $t = 15$ for $Ad = 1.3$ and 64. (c) Gyration radius of agglomerates $R_g(A)/r_p$ as a function of agglomerate size $A$ at $t = 15$ for the cases with $St_k = 5.8$ and $Ad = 13$ (diamonds), 64 (triangles), and 128 (squares). The solid line shows Eq. (12) with $k = 1.75$ and $D_f = 1.57$. (d) An agglomerate produced in the simulation of $St = 5.8$ and $Ad = 64$ with its equivalent sphere with radius of gyration (shaded region).

C. Exponential scaling of early stage agglomerate size

Fig. 4(a) shows the early-stage ($t \leq 20$) distributions of number density of agglomerates as a function of size $A$. These distributions, when scaled by the initial number density of primary particles $n_0$, follows an exponential form (solid lines in Fig. 4(a))

$$\frac{n(A)}{n_0} = \beta \exp\left(-\frac{A}{\kappa}\right),$$

(13)

with the coefficients $\beta$ and $\kappa$ depending on time. This exponential distribution is quite different from the well-known self-preserving size distribution for Brownian nanoparticles [50, 51]. Based on the conservation of total number of primary particles, $\Sigma_1^\infty A \ast n(A) = n_0$, the prefactor $\beta$ can be expressed as $\beta(\kappa) = 2 \cosh(\kappa^{-1}) - 2$. Therefore, the early-stage agglomeration can be characterized using a single scale parameter $\kappa$. In the inset of Fig. 4(a), the number density distributions for cases with $St_k = 5.8$, 12, and 23 and $Ad = 1.3$, 13, and 64 are plotted in a rescaled form, $n(A)/(n_0\beta) \sim A/\kappa$. Except the deviation in tail caused by agglomerates with $n(A)/n_0 < 0.3\%$, the results center around the curve $y = \exp(-x)$, suggesting that the exponential scaling for early-stage agglomeration is valid for inertial particles across a wide range of adhesion force magnitudes.

Now we introduce how to construct the agglomeration kernel that can be applied to Smoluchowski’s theory based on DNS-DEM results. We first look at the strong adhesion case by assuming that particles will stick together upon collision and then show how adhesion influences the sticking probability. For spherical particles, $\Gamma(i,j)$ is given by

$$\Gamma(i,j) = 2\pi R_{ij}^2 |w_r| g(R_{ij}),$$

(14)

where $R_{ij} = r_{p,i} + r_{p,j}$ is the collision radius, $\langle |w_r| \rangle$ is the average radial relative velocity and $g(R_{ij})$ is the radial distribution function at contact. Explicit expressions of these quantities are summarized in [13]. Since turbulence parameters are fixed here, $R_{ij}$, $\langle |w_r| \rangle$ and $g(R_{ij})$ are determined by particle size and $St_k$. For collisions between
FIG. 3. (a), (d) and (g): Temporal evolution of the collision kernel $\Gamma(t)/\Gamma_0$ for cases with $Ad = 0.013$ (circles), 1.3 (left-pointing triangles), 13 (diamonds), 64 (upward triangles), and 128 (squares). (b), (e) and (h): Fraction of particles, $P(A)$, contained in agglomerates of size $A$ at $t = 15$ for $Ad = 1.3$ and 64. (c), (f) and (i): Gyration radius of agglomerates $R_g(A)/r_p$ as a function of agglomerate size $A$ at $t = 15$ for the cases with $Ad = 0.013$ (circles), 1.3 (left-pointing triangles), 13 (diamonds), 64 (upward triangles), and 128 (squares). (a) (b) and (c) are results for $St_k = 0.72$, (d) (e) and (f) are for $St_k = 12$ and (g) (h) and (i) are for $St_k = 23$. The solid lines in (c), (f), and (i) are fits to Eq. (12) with (c) $k = 1.86$ and $D_f = 1.50$, (f) $k = 1.76$, $D_f = 1.56$, and (i) $k = 1.60$, $D_f = 1.64$.

As shown in Fig. 4(b), the kernel $\Gamma(i,j)$ constructed in this way readily reflects the effect of fractal structure of agglomerates on agglomeration process and the PBE in Eq. (10) well reproduces the results of DNS-DEM in Fig. 4(a) when $t \leq 15$ (see also the comparison of $\kappa$ from the two methods in the inset of Fig. 4(b)). At $t = 20$, the distribution of $n(A)/n_0$ from PBE still follows the exponential form given by Eq. (13), however, a non-negligible deviation between the results and those from DNS-DEM is observed. Such deviation can be attributed to two reasons. First, $\Gamma(i,j)$ does not contain information of breakage or rearrangement, which is expected to be significant for large-size agglomerates [26]. Moreover, statistics may also get worse when the total number of agglomerates $\Sigma_1^\infty n(A)$ reduces.

D. Effect of adhesion on growth of agglomerates

When the adhesion is relatively weak, a collision between two particles or agglomerates does not ensure the formation of a larger agglomerate. We then solve PBE using agglomeration kernel $\Gamma_a(i,j)$ with $\Theta$ increasing from 0 to 1 (see Eq. (11)). The evolution of the scale parameter $\kappa$ is shown as solid lines in Fig. 5(a). It is evident from the results that a smaller sticking probability $\Theta$ leads a lower growth rate of agglomerates.

We also plot corresponding results from DNS-DEM simulations with different values of adhesion parameter $Ad$ as...
FIG. 4. (a) Scaled number density $n(A)/n_0$ of agglomerates of size $A$ for the case with $St_k = 5.6$ and $Ad = 64$ at $t = 5$ (circles), $10$ (squares), $15$ (exes), and $20$ (triangles). The solid lines are fits to Eq. (13). Inset: scaled number density $n(A)/n_0$ as a function of $A/\kappa$ for $St_k = 5.8$ (circles), $12$ (triangles), and $23$ (squares). For each $St_k$, results are shown for $Ad = 1.3$ (black), $13$ (blue), and $64$ (red), at $t = 5, 10, 15$. The solid line is exponential scaling $y = \exp(-x)$. (b) $n(A)/n_0$ v.s. $A$ calculated from population balance equations. Legends are the same as panel (a). In the inset of (b), we show the temporal evolution of the scale parameter $\kappa$ from DNS-DEM result (circles) and from PBE (solid line).

data points in Fig. 5(a). For $Ad = 0.013$, $\kappa(t)$ almost keeps constant and is close to the PBE results with sticking probability $\Theta = 0$, indicating that nearly no agglomerates are formed given such a weak adhesion. As $Ad$ increases beyond $\sim 64$, the $\kappa(t)$ curves converge to the PBE result with sticking probability $\Theta = 1$. This strong adhesion case corresponds to the conventional PBE simulations, where the hit-and-stick assumption is made. Our results here suggest that PBE can also simulate the agglomeration process for particles with relatively weak adhesion once the sticking probability $\Theta$ is adopted.

Finally, we determine the value of the sticking probability $\Theta$ in a statistical manner based on our DNS-DEM data. For a given $Ad$, we extract the instantaneous value of the scaling parameter $\kappa(t, Ad)$ from DNS-DEM simulations and map this point out on Fig. 5(a) and find the PBE curve of $\kappa(t, \Theta)$ that the point sits on. This procedure instantaneously correlates $\Theta(t)$ to $Ad$. Then time-averaging is performed to get the sticking probability at this given $Ad$:

$$\Theta(Ad) = \frac{1}{T} \int_0^T \Theta(t) dt.$$ (15)

In Fig. 5(b), we plot $\Theta(Ad)$ for $St_k = 5.8, 12, 23$. With $Ad < 1$, the sticking probability $\Theta$ for any $St_k$ is smaller than $\sim 0.3\%$ and the data points of different $St_k$ are rather scattered. In contrast, when $Ad > 10$, there is an adhesion-controlled regime, in which $\Theta$ is mainly determined by $Ad$. Particularly, the unit sticking probability, $\Theta \approx 1$, which corresponds to the hit-and-stick situation, is achieved when $Ad$ is larger than $\sim 50$.

E. Effect of Reynolds number

In the present work, the computing effort was mainly devoted to resolving the interparticle contacting using DEM. Performing DEM and high-Reynolds-number DNS simultaneously is numerically infeasible for us at this stage. Although we fix the value of Taylor-microscale Reynolds number $Re_\lambda$, the effect of $Re_\lambda$ on the agglomeration can be reasonably speculated based on the Reynolds-number dependence of collision kernels reported in [12, 13, 54] and the sticking probability from this work. Ireland et al. study the particle dynamics in HIT with $Re_\lambda$ ranging from 88 to 597 [54]. They reported that, for inertial particles ($St_k \geq 10$), the radial relative velocity $\langle |w_r| \rangle$ increases strongly with increasing $Re_\lambda$ since the effect of filtering on larger turbulence scales decreases and the particles also carry a memory of more energetic motions as $Re_\lambda$ increases. The RDF, $g(R)$, also increases due to a stronger clustering effect at larger $Re_\lambda$. The combined effect leads to a larger value of the collision kernel at a higher $Re_\lambda$.

On the other hand, the results in Sec. III D implies that the increase of the collision rate does not ensure an enhancement of the agglomeration rate. As shown in Fig. 5(b), the sticking probability is positively correlated to the adhesion parameter $Ad$. For a given value of particle surface energy $\gamma$, the increase of $Re_\lambda$ leads to more energetic collisions between particles, which further leads to a smaller value of $Ad$. Hence, a decrease of the sticking probability $\Theta$ is expected due to the higher relative colliding velocity. A quantitative characterization of competing
FIG. 5. (a) Temporal evolution of the parameter $\kappa$ for DNS-DEM simulation with $St_k = 5.8$ and $Ad = 0.013$ (circles), 1.3 (left-pointing triangles), 13 (diamonds), 64 (upward triangles), 128 (squares), and 256 (axes). The solid lines spanning from light to dark color are results from PBE with the sticking probability $\Theta = 0, 0.2, 0.4, 0.8, \text{and } 1$. (b) Sticking probability $\Theta$ as a function of adhesion parameter $Ad$, determined from Eq. (15), for $St_k = 5.8$ (circles), 12 (triangles), and 23 (diamonds).

effects of the increasing collision rate and the decreasing sticking probability as Taylor-microscale Reynolds number $Re_\lambda$ increases would be of great interest. Moreover, a recent work involving high-Reynolds-number flows reported that the intermittent Lagrangian motion leads to correlated collision events, which further results in a rapid growth of large aggregates. In that case, Smoluchowski’s coagulation kinetics may fail to predict the agglomeration [55]. Therefore, a complete picture of agglomeration should include the role of both the turbulent transport (e.g., vortices and intermittency) and the microphysical mechanisms (particle-level interactions), which is an interesting direction for future research.

IV. CONCLUSIONS

In summary, for adhesive inertial particles suspended in turbulence, we measure both the collision rate, the structure and the size distribution of early-stage agglomerates with varying adhesion. We find that the size distribution follows an exponential scaling $n(A)/n_0 = \beta(\kappa) \exp(-A/\kappa)$. The state of agglomeration thus can be characterized using a single scale parameter $\kappa$. The evolution of $\kappa$ then serves as an indicator for the comparison between DNS-DEM results and the one-dimensional PBE simulation. We show that, by introducing an agglomeration kernel that contains the information of the structure and sticking probability, PBE can be easily extended to simulate the adhesion-controlled agglomeration.

There are several interesting directions for future study. First, the current work focuses on the early-stage agglomeration, where the fractal structure of small-sized agglomerates is well described by the radius of gyration with both prefactor $k$ and fractal dimension $D_f$ insensitive to time and interparticle adhesion. However, collision of large agglomerates can sometimes result in fragmentation into a large number of offspring agglomerates [56], which is not accounted for in the current model. A recent study by Dizaji et al. [57] has shown that even in cases where two colliding agglomerates appear to bounce off of each other, this bouncing process is always accompanied by an exchange of particles between the agglomerates. Computation with larger agglomerates requires investigators to construct kernel functions that contain information about breakage and restructuring [58, 59], and it is unclear to what extent the framework developed here can be extended to situations with large agglomerates. Moreover, the sticking probability $\Theta$ is currently determined by fitting. An explicit expression for the sticking probability $\Theta$ could be constructed based on the statistics on the relative velocity [54, 60] and a sticking-rebound criterion for collisions of adhesive particles [41]. The presence of interparticle electrostatic force can also affect turbulent particle agglomeration. Preliminary results have shown that even the simplest Coulomb interactions play an important role in determining the level of clustering [61, 63]. The interparticle electrostatic force is normally long-ranged and can be either repulsive or attractive. Efficient algorithms, like fast multiple method [64, 65] and $P^3M$ method [63], thus are needed to reduce the computing cost.
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