Coagulation Characteristics of Nano-agglomerates in Free Molecular States under Different Electric Field Intensities

Ting Luo¹, Pengfei Zhang², Dongxu Cheng³, Congcong Li¹, Junyi Shen¹, Mingyue Yang¹, Jie Zhao¹, Kai Zhang¹, Qianyuan Yu²*, Zhandong Shi²*  

¹School of Measurement and Testing Engineering, China Jiliang University, Hangzhou, 310018, China  
²Zhengzhou Tobacco Research Institute of CNTC, Zhengzhou,450001, China  
³China Tobacco Henan Industrial Co., Ltd.  

* Corresponding author. Tel: 138 3880 0777, 183 3996 1526  
E-mail address: yuqianyuan7@163.com(QY. Yu), shizhandong500@126.com(ZD. Shi)

Abstract

The coagulation behavior of nanoparticles in microfluidic chip channels is complex and affected by various external factors, including electric fields, sound fields, and wakes. Among them, electric fields have been confirmed to be the most effective for combining and manipulating particles to create new materials with diverse functionalities. In this work, the coagulation characteristics of nanoparticles in the electric field were studied, and the feasibility of the simplified Smoluchowski’s equation (SE) by Taylor-expansion moment method (TEMOM) when considering non-spherical particle shapes was verified. The results showed that the particle evolution process obtained by introducing fractal dimension was closer to reality. Compared to spherical particles, non-spherical particles are often observed to exhibit potentially stronger coagulation processes, potentially faster aggregation and growth rates, and potentially lower concentrations of surrounding particles. At the same time, due to the increase in collision, coagulation, and fracture times, the dispersed particle system has stronger multi-dispersion, and more new particles of different sizes are formed. Specifically, the concentration of charged non-spherical particles at time $\tau=0.5$ is about 6.25 times lower than that of charged spherical particles. Similarly, at time $\tau=10$, the dispersion of charged non-spherical particles is 700 million times greater than that of charged spherical particles. Furthermore, when charged particles with different fractal dimensions are subjected to the same electric field intensity, their coagulation processes exhibit nearly identical behavior. The results of this study are of great significance for guiding the assembly of nanoparticles.

Keywords: Nanoparticle, Non-spherical, Dielectric particles, Coagulation.
Aerosol refers to suspended particles in the air, and its particle size is generally between 0.01 and 10 microns. Aerosols can be transported and manipulated through microfluidic chip channel. In microfluidic chip channel, the behavior of fine particulate matter and ultrafine particles is exceedingly complex, exhibiting multiple dynamic behaviors such as coagulation, fragmentation, condensation, evaporation, nucleation, deposition, and surface chemical reactions (Frederix, 2016). These dynamic behaviors lead to continuous changes in the size, concentration, spatial distribution, and even chemical composition of the particles, ultimately determining their spatiotemporal dynamics (Zhao, 2007). After the particles collide, they coagulate and assemble into novel materials with specific functions. The functionality of these materials relies on various parameters, including the displacement, orientation, mass concentration, quantity concentration, and other features of the condensed particle string. These parameters can be successfully harnessed to capture, manipulate, and assemble nanoparticles in microfluidic systems (Xiong et al., 2016). Hence, accurately characterizing the dynamic behavior of particles is a crucial aspect of comprehending the characteristics governing their motion. In particle system analysis, akin to the general principles of fluid mechanics, experimental and theoretical methods are employed (Gao, 2020). While the mechanism and process of particle motion phenomena have yet to be fully understood, particle shape can be established by setting conditions in simulation, an approach that cannot be substituted with theoretical analysis. Nevertheless, theoretical analysis
offers a means to overcome the complexity, high cost, and poor repeatability of simulation or experimentation, and enables the identification of internal laws. Therefore, a judicious combination of experimental and theoretical approaches is necessary to tackle the coagulation issue of particles in microfluidics (Xu et al., 2021).

Particle coagulation occurs when two or more particles collide and bond to form a larger particle. Several methods have been developed to solve the equation describing this process, such as the partition method, Monte Carlo method and moment method. The partition method discretizes the particle size distribution curve into n intervals and assumes a uniform distribution function for the particle size in each interval. A balance equation is established for the distribution function of a particle attribute in each interval and solved simultaneously to obtain the temporal evolution of the distribution function of that attribute (Gelbard et al., 1980). The Monte Carlo method simulates directly the Boltzmann equation of gas dynamics (Shen, 2006).

The moment method describes the evolution of the moment values of the particle-scale distribution function. Lee (1983) first applied the moment method to solve the Brownian coagulation problem of particles in the continuous region, and then investigated how the coagulation rate of polydisperse particles varies in different regimes (free molecular, transition, and continuum). The results show that polydisperse particles have higher coagulation efficiency than monodisperse particles (Lee and Chen, 1984). Lee et al. (1997) also studied the variation of
the particle size distribution function with time under the condition of low Knudsen number and
in the whole interval (Park et al., 1997). However, the traditional method of moments has
limitations as it assumes the initial particle scale distribution function, such as log-normal or
gamma distribution, which may not necessarily apply to real particles. Yu et al. (2008) proposed
a new form of moment equation that does not require assumptions about the initial distribution of
particles, making it more generalizable. In his doctoral dissertation, he elaborated TEMOM and
deduced the Taylor-expansion moment equation of particles in the free molecular region,
continuous region, and whole region. This method was applied to verify the Brownian
coagulation problem of full-interval particles (Yu et al., 2011) and the particle fragmentation
problem (Yu and Zhang, 2012), showing better computational efficiency and accuracy compared
to other methods. Further analysis and research on the coagulation problem of each interval (Lin
and Chen, 2013) have been carried out using this method, with studies also investigating
polymerization and collision in high concentrations for Brownian coagulation (Wang et al., 2021).

The conductive properties of nanoparticle surfaces can be categorized into three types:
insulation, dielectric, and conductivity (Zhao et al., 2018). In the electrodynamic study of
microparticles (Zhang et al., 2010), spherical particles in a uniform electric field can be assumed
to be equivalent to point electric dipoles due to the thin electric double layer. Gangwal et al.
(2008) from Massachusetts Institute of Technology (MIT) investigated the effects of particle
shape and electrical conductivity on induced electroosmotic flow and electrophoresis in particle multiphase flow. Touvia (2013) developed an ideal polarizable, rigid-body nanoparticle translation and angular velocity relationship with the applied electric field for any thickness and shape of the electric double layer, based on fluid continuity and weak electric field assumptions. Yang and Lin (2021) proposed the general dynamics equation of particles in micro-nano two-phase turbulence and analyzed the unsealed source term caused by turbulence pulsation contained in the equation. They reviewed the effects of turbulent pulsation on particle nucleation, growth, coagulation, and fragmentation, as well as studying the distribution and deposition of cylindrical nanoparticles in turbulent flow (Lin et al., 2021).

However, the current dynamics model equation of particulate matter still has room for improvement. For instance, in the study of particle coagulation behavior, the assumption of spherical particles is often made, which does not reflect reality. Only a few researchers, such as (Yu et al., 2016; Yu et al., 2017a; Yu et al., 2017b) have developed dynamics models for non-spherical particles based on the coagulation model in continuous and near-continuous regions. Therefore, building upon prior research, this study introduces the concept of fractal dimension to simulate the motion state of non-spherical particles. Drawing an analogy to magnetic fields, the electric field is extended to a non-spherical mode, and an electric field condensation moment
equation model is obtained. The influence of different electric field strengths on the condensation of non-spherical particles is also examined.

2 Theoretical Model

This chapter provides an overview of theoretical models for particle coagulation. Specifically, it covers models for coagulation in the free molecular region of both spherical and non-spherical particles, with a focus on their behavior under the influence of magnetic and electric fields.

2.1 Free-Region Brownian Coagulation Model

2.1.1 SE Equation for Uncharged Spherical Particles

In 1917, Smoluchowski proposed the mathematical expression for the particle number concentration in a polydisperse system during the process of particle coagulation, based on the classical Smoluchowski mean field theory (Smoluchowski, 1918). However, the expression was in a discrete form. Later, in 1928, Muller built on Smoluchowski’s work and derived the calculus form of the problem. Presently, research on discrete systems is based on the Muller equation, which is represented as (Müller, 1928):

\[
\frac{\partial n(v,t)}{\partial t} = \frac{1}{2} \int_0^v \beta(v_1,v-v_1)n(v_1,t)n(v-v_1,t)dv_1 - n(v,t) \int_0^\infty \beta(v_1,v)n(v_1,t)dv_1
\]  

where \(n(v,t)\) is the number concentration of particles with volume \(v\) at time \(t\), and \(\beta(v_1,v)\) is the collision kernel function of two particles with volumes \(v\) and \(v_1\). Despite the framework of the Stokes-Einstein theory, no one has yet provided an analytical solution to this equation without any assumptions. The crux of solving this problem lies in converting the calculus form of the SE
equation to the moment equation in differential form. By multiplying the SE equation by \( v^k \) and integrating it with \( v \), we can obtain the time variation of each order moment, as follows:

\[
\frac{dm_k}{dt} = \frac{1}{2} \int_0^\infty \int_0^\infty [(v + v_1)^k - v^k - v_1^k] \beta(v, v_1) n(v, t) n(v_1, t) dv dv_1
\]

(2)

Among them, the higher-order moments are defined as follows:

\[
m_k = \int_0^\infty v^k n(v) dv
\]

(3)

In the Eq. (3), take the Taylor expansion of \( v^k \) at \((v = u)\) and the first three terms are taken, and reveals that any moment can be represented as a linear combination of \( m_0 \), \( m_1 \), and \( m_2 \). Specifically, \( m_0 \), \( m_1 \), and \( m_2 \) correspond to the zeroth, first, and second-order moments, respectively.

In the Brownian coagulated TEMOM model of free-region proposed by (Lee et al., 1997; Yu and Lin, 2010), the mean particle volume is defined as \( \bar{\nu} = m_1 / m_r \). This is chosen as the central expansion point of the third-order Taylor expansion. The literature describes the principle of selecting expansion points and the issue of accuracy. This method enables the direct calculation of the TEMOM moment method expression from the TEMOM model:

\[
\begin{align*}
\frac{dm_0}{dt} &= \sqrt{2} B_1 (65 m_2^2 m_0^{23/6} - 1210 m_2 m_1^2 m_0^{17/6} - 9223 m_1^4 m_0^{11/6}) / 5184 m_1^{23/6} \\
\frac{dm_1}{dt} &= 0 \\
\frac{dm_2}{dt} &= -\sqrt{2} B_1 (701 m_2^2 m_0^{11/6} - 4210 m_2 m_1^2 m_0^{5/6} - 6859 m_1^4 m_0^{-1/6}) / 2592 m_1^{11/6}
\end{align*}
\]

(4)

2.1.2 SE Equation of Uncharged Spherical Particles Considering Fractal Dimension
The previous studies on nanoparticles assume that they are spherical. However, in reality, most nanoparticles exhibit irregular shapes, as depicted in Fig. 1, which shows the microstructure of the particles obtained by scanning electron microscopy (SEM) observation. This creates a certain deviation between the theoretical research results and the actual scenario. Hence, further theoretical research on this problem is imperative. It is necessary to consider this factor in the coagulation model and optimize it further by incorporating the concept of fractal dimension.

When taking into account the fractal dimension $D_f$, the collision kernel function of the Brownian coagulation mechanism can be expressed as:

$$
\beta^D_{dep,FM} = B^D_{i} \eta_{dep,FM} \left( \frac{1}{v} + \frac{1}{v_i} \right)^{1/2} \left( v^{1/\lambda} + v_i^{1/\lambda} \right)^2
$$

(5)

$$
B^D_i = \frac{6k_b T c}{\rho} \left( \frac{3}{4\pi} \right)^{1/2} \frac{r_0^{1/2}}{v_i^{1/2}}
$$

(6)

where the uniformity parameter is denoted by $\lambda$, which is calculated as $\lambda = 2/D_f - 1/2$. The radius of gyration of the first particle in a pair of coagulated particles with volumes $v$ and $v_i$ is represented by $r_0$. 

![Particle microstructure diagram obtained by SEM observation (Zhang et al., 2020a)](image)
Expand \((1/v + 1/v_i)\) in Eq. (5) using a bivariate Taylor series and retaining the first three terms, and then substituting it into Eq. (2) along with Eq. (5), yields:

\[
\begin{aligned}
\frac{dm_0}{dt} &= B_{\text{dep, fm}}^D \eta_{\text{dep, fm}} \int (2\sqrt{2}v + 2\sqrt{2}v_i + \sqrt{2}v_i^2 - \sqrt{2}v_i) \\
\frac{dm_1}{dt} &= 0 \\
\frac{dm_2}{dt} &= B_{\text{dep, fm}}^D \eta_{\text{dep, fm}} \int (2\sqrt{2}v + 2\sqrt{2}v_i + \sqrt{2}v_i^2 - \sqrt{2}v_i) \\
\end{aligned}
\]  

(7)

By defining the mean particle volume as \(\bar{v} = m_1/m_0\) and substituting it into Eq. (7), the TEMOM moment method expression that considers the particle shape can be obtained:

\[
\begin{aligned}
\frac{dm_0}{dt} &= -\frac{\sqrt{2}B_{\text{dep, fm}}^D \eta_{\text{dep, fm}}}{64}(\epsilon_1 \delta_1 + \epsilon_2 \delta_2 + \epsilon_3 \delta_3) \\
\frac{dm_1}{dt} &= 0 \\
\frac{dm_2}{dt} &= -\frac{3\sqrt{2}B_{\text{dep, fm}}^D \eta_{\text{dep, fm}}}{32}(\xi_1 \phi_1 + \xi_2 \phi_2 + \xi_3 \phi_3)
\end{aligned}
\]  

(8)

where:

\[
\begin{aligned}
\epsilon_1 &= 1 - 24D_{j^{-1}} + 70D_{j^{-1}} - 48D_{j^{-1}} + 16D_{j^{-1}}^2 + 2D_{j^{-1}}^2, \\
\epsilon_2 &= 54 - 144D_{j^{-1}} + 52D_{j^{-1}}^2 + 96D_{j^{-1}} - 32D_{j^{-1}}^4, \\
\epsilon_3 &= 73 + 168D_{j^{-1}} - 122D_{j^{-1}} - 48D_{j^{-1}} + 16D_{j^{-1}}^4, \\
\delta_1 &= m_{-1}^{(4-D_{j^{-1}})^2} m_{-1}^{(4-D_{j^{-1}})^2} m_{-1}^{(4-D_{j^{-1}})^2} m_{-1}^{(4-D_{j^{-1}})^2} m_{-1}^{(4-D_{j^{-1}})^2}, \\
\delta_2 &= m_{-1}^{(4-D_{j^{-1}})^2} m_{-1}^{(4-D_{j^{-1}})^2} m_{-1}^{(4-D_{j^{-1}})^2} m_{-1}^{(4-D_{j^{-1}})^2} m_{-1}^{(4-D_{j^{-1}})^2}, \\
\delta_3 &= m_{-1}^{(4-D_{j^{-1}})^2} m_{-1}^{(4-D_{j^{-1}})^2} m_{-1}^{(4-D_{j^{-1}})^2} m_{-1}^{(4-D_{j^{-1}})^2} m_{-1}^{(4-D_{j^{-1}})^2}, \\
\xi_1 &= 1 + 16/3D_{j^{-1}} + 10D_{j^{-1}} / 3 - 16D_{j^{-1}} / 3 - 16D_{j^{-1}} / 3, \\
\xi_2 &= 2/3 - 32D_{j^{-1}} / 3 - 32D_{j^{-1}} / 3, \\
\end{aligned}
\]
\[ \xi_i = -133/3 + 80D_f^{-1}/3 + 202D_f^{-2}/3 - 16D_f^{-3}/3 - 16D_f^{-4}/3, \]

\[ \varphi_i = \frac{m_0^{(4+D_f)\geq 0}}{m_1^{(4-D_f)\geq 0} m_2^{(6-D_f)\geq 0}}, \quad \varphi_s = \frac{m_0^{(4+D_f)\geq 0}}{m_1^{(4-D_f)\geq 0} m_2^{(6-D_f)\geq 0}}, \quad \varphi_s = \frac{m_0^{(4-D_f)\geq 0}}{m_1^{(4+D_f)\geq 0} m_2^{(6+D_f)\geq 0}}, \]

In the experiment conducted by (Yamaguchi et al., 2011), the microscopic coagulation structure of NaCl particles was observed using a projection electron microscope. The results showed that when the fractal dimension \( D_f = 3 \), the coagulation diameter was consistent with the equivalent diameter, and the coagulated particles could be regarded as spheres.

When \( D_f = 3 \) is brought into Eq. (8), it coincides with Eq. (4), thereby validating the accuracy of the deduced calculation method that considers particle shape. The model time is dimensionless and is obtained by using the equation \( \tau = B_1 t \), which is a simplified processing method based on the research of Barrett & Webb in 1998. Our model code produces results that are consistent with those obtained using TEMOM, demonstrating the reliability of our programming and the rationality of our model in considering fractal dimensions.

### 2.2 Free-Region Electrocoagulation Model

#### 2.2.1 SE Equation for Charged Spherical Particles

When two particles are nearby, the interparticle dielectric force increases rapidly, making short-range physical and chemical interactions, such as van der Waals forces and the double-layer effect, negligible in polar particle coagulation models. Compared to ferromagnetic particle coagulation, the impact of van der Waals and hydrodynamic force is insignificant, and therefore,
it is feasible to consider only magnetic force in particle coagulation. Additionally, the electric field force can also be considered separately.

Friedlander and Kumar proposed the collision kernel function between spherical magnetic dipoles, as presented in their work (Friedlander and Marlow, 1977; Kumar and Biswas, 2005):

\[
\beta_{\text{mig, fm}} = \frac{1}{6\pi \epsilon_0} \left[ \frac{\pi \chi_{mi} \chi_{mj} B^2}{\mu_0} \right]^{1/3} (k_B T)^{1/6} \frac{1}{\rho_p^{1/2}} \left( \frac{1}{d_i} + \frac{1}{d_j} \right)^{1/2} (d_i + d_j)
\]  

(9)

By converting the diameter to volume using the formula \( v = \pi d^3 / 6 \) and \( v_1 = \pi d_1^3 / 6 \), and through the application of electromagnetic analogy, we can obtain the collision kernel function between electric dipoles in the electric field:

\[
\beta_{\text{dep, FM}} = B_i \eta_{\text{dep, fm}} \left( \frac{1}{v} + \frac{1}{v_1} \right)^{1/2} \left( v^{2/3} v_1^{1/3} + v^{1/3} v_1^{2/3} \right)
\]  

(10)

where: \( B_i = (3/4\pi)^{1/6} (6k_B T / \rho)^{1/2} \), \( k_B \) is Boltzmann constant, \( T \) is the gas temperature, \( \rho \) is the particle density, \( \eta_{\text{dep, fm}} = (3^{-1/6} \cdot 2^{1/3} / 3 \cdot 6^{1/2} / 6)(k_B T)^{1/6} [\epsilon_v / \epsilon_0 - 1] (\epsilon_i / \epsilon_0 - 1) E^2 \), \( \epsilon_0 \) is the vacuum permittivity, \( \epsilon_{i, j} \) is the absolute permittivity of the particles, and \( E \) is the applied electric field strength.

To facilitate the transformation of the scale spectrum space on the right-hand side of the equation to the moment variable space, Eq. (10) is first transformed into the following form:

\[
\beta_{\text{dep, FM}} = B_i \eta_{\text{dep, fm}} (v + v_1)^{1/2} (v^{1/6} v_1^{-1/6} + v^{-1/6} v_1^{1/6})
\]  

(11)

Expanding \( (v + v_1)^{1/2} \) in Eq. (11) using the Taylor series at \( (v = u, v_1 = u) \) and combining it with Eq.(2) and Eq. (11), we obtain the TEMOM first-order ordinary differential equation system for
numerical solution (Zhang et al., 2020b). Here, the average particle volume is denoted by
\[ \bar{v} = \frac{m_1}{m_0}. \]

\[
\begin{align*}
\frac{dm_0}{dt} &= -\sqrt{2} B_I n_{dep, frw} \left(11m_2^2m_0^2 - 382m_2m_0^2m_0 + 10739m_1^4\right)m_0^{3/2} \\
&= -\frac{10368m_0^{3/2}}{10368m_0^{3/2}} \\
\frac{dm_1}{dt} &= 0 \\
\frac{dm_2}{dt} &= \sqrt{2} B_I n_{dep, frw} \left(709m_2^2m_0^2 - 6242m_2m_0^2m_0 - 4835m_1^4\right) \\
&= -\frac{5184m_1^{3/2}m_0^{1/2}}{5184m_1^{3/2}m_0^{1/2}}
\end{align*}
\]

(12)

2.2.2 SE Equation for Charged Non-spherical Particles

Fig. 2 illustrates the collision and coagulation of non-spherical nanoparticles under the
influence of an electric field. As the electric field intensity is applied on both sides of the space,
the nanoparticles undergo continuous collision and coagulation. In microchannels filled with
electrolytes, non-spherical nanoparticles are vertically released and exposed to an applied electric
field, resulting in polarization and the acquisition of an electrical charge that modifies the
surrounding electric field. Consequently, the disturbance in the electric field induces neighboring
particles to polarize, leading to a deviation from their original freely diffusing state and resulting
in the formation of particle agglomerates with a certain degree of regularity.

Fig. 2. Schematic diagram of collision and coagulation of non-spherical nanoparticles in an
electric field
Incorporating the fractal dimension, the collision kernel function for particle coagulation under
the influence of an electric field can be expressed as:

\[ \beta_{\text{dep,FM}} = B_i^D \eta_{\text{dep, fm}} \left( \frac{1}{v} + \frac{1}{v_1} \right)^{1/2} \left( v^{1/2}_B v^{1/2}_D f_B + v^{1/2}_D v^{1/2}_F f_F \right) \tag{13} \]

To simplify the calculation, we can derive a simplified version of Eq. (13):

\[ \beta_{\text{dep,FM}} = B_i^D \eta_{\text{dep, fm}} \left( v + v_1 \right)^{1/2} \left( \frac{2}{v^{1/2}_B v^{1/2}_D f_B} + v^{1/2}_D v^{1/2}_F f_F \right) \tag{14} \]

To facilitate a direct solution, the variable part in Eq. (14) can be expanded in a Taylor series
with \( v = u \), and \( v_1 = u \) as the center, with the first three terms retained. By combining this with Eq.
(2) and Eq. (13), a third-order moment expansion using the TEMOM method is performed. This
leads to an expression that can be directly solved using the TEMOM moment method:

\[
\begin{align*}
\frac{d m_0}{dt} &= -\frac{\sqrt{2} B_i^D \eta_{\text{dep, fm}}}{128} \left( \epsilon_1 \delta_1 + \epsilon_2 \delta_2 + \epsilon_3 \delta_3 \right) \\
\frac{d m_1}{dt} &= 0 \\
\frac{d m_2}{dt} &= -\frac{3\sqrt{2} B_i^D \eta_{\text{dep, fm}}}{64} \left( \xi_1 \varphi_1 + \xi_2 \varphi_2 + \xi_3 \varphi_3 \right)
\end{align*}
\tag{15}
\]

where:

\[ \epsilon_1 = 1 - 36D_f^{-1} + 182D_f^{-2} - 288D_f^{-3} + 128D_f^{-4}, \]

\[ \epsilon_2 = 54 - 216D_f^{-1} - 44D_f^{-2} + 576D_f^{-3} - 256D_f^{-4}, \]
3 Results and Discussion

3.1 Comparison of Brownian Coagulation of Uncharged Non-spherical and Spherical Particles in Free Region

Furthermore, a comparison was made between the Brownian coagulation moments of uncharged spherical particles and non-spherical particles in the free molecular region. The temporal evolution of the zero-order and second-order moments of the two-particle shapes is presented in Fig. 3. Specifically, Fig. 3(a) depicts the change in the number concentration of particles in the system. As shown, non-spherical particles exhibit a lower particle number concentration than their spherical counterparts within the same timeframe. For instance, at $\tau=2$, the particle number concentration of spherical particles is approximately 1.64 times higher than that of non-spherical particles. This observation illustrates the impact of particle shape on the collision and condensation process in the system, thereby suggesting the need to account for particle shape when studying the coagulation process. On the other hand, Fig. 3(b) demonstrates the variation in the polydispersity of particles in the system. The second-order moment dispersion of particle coagulation considering particle shape is stronger when compared to the Brownian coagulation of spherical particles. The dispersion of non-spherical particles is nearly five times
stronger than that of spherical particles at $\tau=10$, indicating that the rate of particle collision, coagulation, and fragmentation in the actual system is also faster in generating new particles of different diameters. Hence, it can be inferred that the introduction of the fractal dimension is necessary for the accurate calculation of the coagulation process.

Fig. 3. Evolution of two different moments with time $\tau$ for non-sphere and sphere under Brownian coagulation (a) zero order moment (b) second-order moment

Fig. 4 depicts a comparative analysis of the temporal evolution relationship between zero-order and second-order moments at varying fractal dimensions. As illustrated in Fig. 4(a), a decrease in fractal dimension leads to a steeper decline in particle number concentration. Additionally, Fig. 4(b) reveals the impact of fractal dimension on the second-order moment. Specifically, as the fractal dimension decreases, the polydispersity of the system increases, with a shorter time needed to achieve the same level of polydispersity. This trend aligns with the understanding that lower fractal dimensions correspond to looser shapes and structures, thereby providing more opportunities for collisions, coagulations, and ruptures.
3.2 Coagulation Contrast of Charged Non-spherical and Spherical Particles in Electric Field

To compare the coagulation behavior of charged spherical and non-spherical particles in the free molecular region, we examined the zero-order and second-order moments of two different particle shapes as a function of time $\tau$ are shown in Fig. 5. Fig. 5(a) indicates that the coagulation of non-spherical particles leads to a lower particle number concentration compared to the electrocoagulation of charged spherical particles within the same period. Furthermore, the particle number concentration decreases at a faster rate in the case of non-spherical particles, as evidenced by the significant difference in concentration already observed at $\tau=0.5$. These findings suggest that particle collision and coagulation are more pronounced in systems containing non-spherical particles. Similarly, Fig. 5(b) illustrates that non-spherical particles exhibit stronger dispersion in particle volume compared to charged spherical particles, with the electric field also enhancing the dispersion effect. Specifically, at $\tau=10$, the dispersion of charged non-spherical particles is about 700 million times stronger than that of spherical particles.
3.3 Coagulation Characteristics of Charged Non-spherical in Electric Field with Different Fractal Parameters and Electric Field Strengths

In this section, we focus on the coagulation behavior of charged non-spherical particles in the electric field. The ability to combine and manipulate particles in such fields has made it possible to synthesize novel materials with diverse functionalities. Notably, the morphologies of the assembled particles differ depending on the shapes of the individual components, with the electric field intensity serving as the primary modifiable factor. Consequently, we concentrate on investigating the impact of the fractal dimension and electric field intensity on the coagulation dynamics.

3.3.1 Different Fractal Parameters

Simultaneously, when considering the particle shape, the evolution relationship of different fractal dimensions to the zero-order and second-order moments with time $\tau$ is also compared, as graphically depicted in Fig. 6.

As illustrated in Fig. 6(a), the decrease of fractal dimension in various particles results in a decline in particle number concentration at the same time, indicating particle coagulation. This
correlation implies that a lower fractal dimension is linked with more intense collision and coagulation in the system. Furthermore, Fig. 6(b) illustrates that as the fractal dimension decreases, the system dispersion rises, signifying that the collision, coagulation, and breakup of particles accelerate, thus producing new particles of varying sizes. The nanoparticles' fractal parameters were adjusted without modifying the dielectric nanoparticle's electric field strength or polar conditions. When the fractal parameters were reduced, the particle number concentration increased, and as the coagulated particle concentration rose, the number of particles decreased. This observation suggests that smaller fractal parameters lead to looser particle coagulation and more vigorous particle collision and solidification processes. This effect can expedite particle coagulation and expansion, leading to a quicker speed and fewer particles. By comparing the two approaches, we concluded that the distribution of particle diameter values considering particle shape was broader than that of spherical particle coagulation processes. This conclusion aligns entirely with the findings of Heine and Pratsinis (Heine and Pratsinis, 2007).

Fig. 6. Evolution of two different moments with time $\tau$ under different fractal dimensions
(a) zero order moment (b) second-order moment

3.3.2 Different Electric Field Strengths
Through the employment of the free zone electrocoagulation and TEMOM moment method, it has been determined that the rate and pattern of particle coagulation are significantly influenced by the electric field intensity, as evidenced by the parameter $\eta_{dep,m}$ derived from the expansion:

$$\eta_{dep,m} = \left(3^{-3/2} \cdot 2^{1/2} / 3 \cdot 6^{1/2} / 6\right) (k_c T)^{1/2} [e_C (\varepsilon_C / \varepsilon_s - 1) (\varepsilon_C / \varepsilon_s - 1) \varepsilon_C]^{1/2}$$

(16)

Fig. 6(a) and (b) exhibit the temporal evolution correlation of zero-order and second-order moments, with varying electric field intensities of 1 kV/m, 3 kV/m, and 5 kV/m, when the fractal dimension is set at 2.1. It is noteworthy that the zero-order and second-order moments of spherical particles display a comparable tendency for different electric field intensities.

![Fig. 7. Evolution of two different moments with time $\tau$ under different electric field strengths](image)

(a) zero-order moment (b) second-order moment

As illustrated in Fig. 7(a), the particle number concentration declines as the electric field intensity increases under distinct electric field intensifiers during the same time interval. Specifically, for each 2 kV/m rise in electric field strength at $\tau=0.1$, the particle number concentration decreases approximately threefold. This observation suggests that the process of
particle collision and coagulation is intensified as the electric field strength increases. Fig. 7(b) also indicates that the system dispersion is enhanced with the increase of electric field intensity during the same time interval, implying that particle collision, coagulation, and rupture produce novel particles with varied diameters at a quicker pace.

With a fixed fractal dimension, the shape of dielectric nanoparticles remains constant while their electric field intensity varies. As the electric field intensity increases, the condensation particle concentration also increases, leading to a reduction in the particle count. This suggests that greater particle coagulation and a more relaxed structure contribute to an intensified particle collision process. Moreover, the higher the electric field intensity, the more substantial the process of particle coagulation and the accelerated decline in particle number concentration. Furthermore, the collision, coagulation, and rupture of particles generate novel particles with distinct diameters at a more rapid rate, thereby enhancing the multi-dispersion of the system.

4 Conclusion

In this study, the collision frequency function of charged particle collision and coagulation was derived from the collision of particles in a magnetic field and the kernel function using the analogy method. The Taylor expansion method was utilized to handle the intricate integer and non-integer terms of the SE transformation equation. This approach enabled the development of the TEMOM model, which describes the coagulation of particles in the free molecular region and
under an electric field with varying particle shapes. The validity and accuracy of the TEMOM model were assessed by comparing the numerical results at discrete points and verifying the numerical results of particle evolution over continuous time scales. This model is expected to find broad application in the large-scale computation of complex multiphase systems. In summary, the TEMOM model developed in this study demonstrates a robust framework for particle coagulation modeling:

1. Compared to spherical particles, non-spherical particles exhibit lower particle number concentrations and stronger dispersion under the influence of both magnetic and electric fields. These findings suggest that non-spherical particles undergo more intense collision and coagulation processes, leading to a faster formation of particles with different diameters, once the particle shape is taken into account.

2. For charged particles, the motion is accelerated by the electric field. As the intensity of the electric field increases, the collision and solidification process of particles within the system become more intense, resulting in accelerated particle growth and reduced concentration of surrounding particles.

3. The smaller the fractal dimension of charged particles, the more intense the movement of particles in the system, the more collision, condensation, and fragmentation times, the more particles of different sizes are formed, and the more polydisperse the system is enhanced.

In the field of air quality monitoring, this study holds significant importance. It focuses on the movement of charged particles in electric fields and the collision and coagulation processes among particles, contributing to a better understanding of the behavior of nanoparticles in the
atmosphere. Such insights have practical applications in improving air quality monitoring technologies, assessing urban environments, and predicting changes in air quality. In this study, when dealing with particulate matter, it is common to treat it as a continuous phase, considering only the effects of the flow field on the particles while neglecting the influence of particles on the flow field. This assumption is reasonable for most practical environmental problems. However, in certain special cases, such as particle flows generated within swirling combustion chambers, the impact of particles on the air flow cannot be ignored. Therefore, a more comprehensive study of the coupling mechanism between particles and the flow field is needed, leading to the development of bidirectional coupled numerical models.

ACKNOWLEDGMENTS

This work was supported by the National Natural Science Foundation of China (No. 11472260).

DISCLAIMER

There are no conflicts of interest to declare.

REFERENCES


ACCEPTED MANUSCRIPT

Technology 3, 327-334. https://doi.org/10.1080/02786828408959020


flow systems. Acta Aerodynamica Sinica 39, 109-120. https://doi.org/10.7638/kqdlxxb-
2021.0030

2009.08.006

for Particles in Brownian Motion Aerosol Science and Technology 42, 705-713.
https://doi.org/10.1080/02786820802232972

for the nanoparticle coagulation in the entire size regime due to Brownian motion Journal of

Yu, M., Lin, J., Seipenbusch, M., Cao, J. (2017a). Verification of size-resolved population
balance modeling for engineered nanoparticles under high concentration Chemical Engineering

https://doi.org/10.1016/j.apm.2016.01.009

Yu, M., Liu, Y., Koivisto, A.J. (2017b). An Efficient Algorithm Scheme for Implementing the


Zhao, H. (2007). Stochastic solution of population balance modeling and the research on high-efficiency removal of particulate matter from coal combustion, Huazhong University of Science and Technology.

Engineering 140, 091302. https://doi.org/10.1115/1.4039709
Table 1. List of variables used in the equations

<table>
<thead>
<tr>
<th>Variable</th>
<th>Definition</th>
</tr>
</thead>
<tbody>
<tr>
<td>$t$</td>
<td>Time</td>
</tr>
<tr>
<td>$v$</td>
<td>Volume</td>
</tr>
<tr>
<td>$\bar{v}$</td>
<td>The mean particle volume</td>
</tr>
<tr>
<td>$D_f$</td>
<td>The fractal dimension</td>
</tr>
<tr>
<td>$\lambda$</td>
<td>The uniformity parameter</td>
</tr>
<tr>
<td>$k_b$</td>
<td>Boltzmann constants</td>
</tr>
<tr>
<td>$T$</td>
<td>The gas temperature</td>
</tr>
<tr>
<td>$\rho$</td>
<td>The particle density</td>
</tr>
<tr>
<td>$\varepsilon_0$</td>
<td>The vacuum permittivity</td>
</tr>
<tr>
<td>$\varepsilon_{i,j}$</td>
<td>The absolute permittivity of the particles</td>
</tr>
<tr>
<td>$E$</td>
<td>The applied electric field strength</td>
</tr>
</tbody>
</table>