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Verification of size-resolved population balance modeling for engineered nanoparticles under high concentration

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HIGHLIGHTS

- The feasibility of PBM for characterizing size-resolved fractal-like ENPs under high concentration was studied.
- The crucial factors affecting the accuracy of the PBM for ENP were revealed.
- The Thajudeen et al.'s aggregate function was verified as the most accurate function as the PBM was implemented.

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ABSTRACT

Concerns have increased regarding the efficacy of population balance modeling (PBM) for determining the size-resolved behavior of engineered nanoparticles (ENPs) in chemical reactors, flames and workplaces. For the first time, we used a well-designed experiment to verify the feasibility of PBM and crucial factors affecting the accuracy of this method when size-resolved behavior was primarily concerned. The dynamic processes were designed to maximally represent high concentration involving aggregate production, deposition, coagulation, and transport. A population balance equation corresponding to the physical changes in an experiment was established and was further solved using the highly accurate moving sectional method. We verified four representative aggregate collision rate functions, namely the modified Fuchs collision rate function, Dahneke's collision function, harmonic mean collision function, and aggregate function newly developed by Thajudeen et al. (Aerosol Sci. Technol. 46 (2012) 1174–1186). The PBM implemented using the Thajudeen et al.'s aggregate function revealed highest agreement between the simulation and measurement. We observed both fractal dimension and primary particle diameter have apparent effects on the accuracy of PBM, indicating that both are key parameters in the implementation of PBM, whereas the pre-exponential factor only slightly affects the accuracy of PBM. The PBM with constant primary particle size, fractal dimension, and pre-exponential factor was finally verified as a reliable method for studying size-resolved evolution of ENPs over time.

1. Introduction

With the development of modern nanoparticle synthesis engineering [1–3], the numerical method for characterizing nanoparticle evolution from gas phase to product undergoes rapid development. Such particles are usually fractal-like aggregates composed of much smaller primary particles, with their diameters varying from approximately ~1 nm to ~1 μm [1,3]. From particle formation to grew particle, besides external processes such as convection and diffusion, there are several internal dynamics processes, including coagulation, production with nucleation mechanism or chemical react mechanism, sintering, and deposition [4]. The most suitable method describing this type of processes is population balance modeling (PBM), which is expected to capture the evolution of nanoparticle size distribution (PSD) through solving the coupled Navier-Stokes equations and population balance equation (PBE) [2,5–7]. However, there are many input parameters and functions which need to be specified with experiment or experiences in advance when implementing the PBM, such as coagulation kernel function and properties related

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Nomenclature

d_f	Aggregate characteristic diameter
D_f	Fractal dimension of aggregates
k_f	The pre-exponential factor
d_{p0}	Primary particle diameter
d_{mv}	Mass equivalent diameter of the aggregate
$n(v,t)$	Particle number concentration with respect to particle volume v
v_{mv}	Mass equivalent volume of aggregates
d_m	Electric mobility diameter
$C(d_m)$	Slip correction for the aggregate mobility diameter
F_{in}, F_{out}	Air flow rate
V	Chamber volume
s	Constant production rate
v_{dv}, v_{du}, v_{dd}	Deposition velocities
A_v, A_u, A_d	Wall areas
u^*	Friction velocity
D_i	Diffusion coefficient
m_i	Particle mass
k_b	Boltzmann constant
T	Temperature
Kn	Knudsen number
c_u	CO_2 volume concentration

Greek symbols

λ	Mean free path of the gas
β	Coagulation kernel
$\in_d$	Deposition loss rate
v	Gas kinetic viscosity
μ	Air viscosity
σ_g	Geometric standard deviation

to nanoparticle morphology [7–10]. This results in some uncertain of the PBM. In fact, it is unclear to what extent PBM agrees with the experiment under high number concentration involving multiple physical changes, such as production release, coagulation, deposition, and transport. For the first time, this work was designed to verify the efficacy of PBM for grasping detailed PSD during nanoparticle evolution, and attempt to make clear crucial factors affecting the accuracy of this method. The dynamic processes were designed to maximally represent realistic high number concentration condition involving aggregate production, deposition, coagulation, and transport. To achieve the quantitative comparison of the evolving PSD between the PBM and experiment, the calculation for Navier-Stokes equation was not involved, which makes the comparison affected by uncertain as little as possible.

The Smoluchowski mean-field theory has been the fundamental theory for studying aerosol dynamics [11]. The key to this theory is its general but powerful governing equation [i.e., the PBE] [4]. The PBE is an integral–differential Fokker–Planck equation that considers the particle number intensity, and it can provide details of the evolution of the aerosol size distribution and statistical quantities, such as the total particle number and volume concentration, over time. Because of its inherent advantage of coupling to computational fluid dynamics within an Eulerian–Eulerian framework, the PBE and relevant modeling method (i.e., PBM) play an increasingly crucial role in both environmental and chemical engineering [12,13]. However, how to validate PBM by using a suitable experimental method remains unclear, particularly for tracing the evolution of size-resolved aggregate distribution over time in engineering condition. Currently, the most common method to validate a novel PBM model involves selecting a more accurate model as the reference. Notably, almost all studied PBM models, including the method of moments (MOM) and Monte Carlo, have used the sectional method (SM) as a reference because of its higher accuracy [7,14,15].

A major limitation of the aforementioned verification is that PBM is validated mathematically rather than physically. Thus, whether PBM provides realistic information on PSD evolution for fractal-like ENPs under high concentration and the extent to which PBM agrees with real physics are both unclear. In addition, the key factors affecting the reliability of PBM under realistic high emission scenarios are unknown. A comparative study of experiments and models under realistic scenarios condition typically has great potential to address these concerns. However, experiments with information identical to PBM are not easily practicable. The main difficulty is because of joint dynamical processes for aerosols that cannot be separated during the experimental measurements [8]. Under realistic high concentration, these aerosol dynamical processes include aggregate release or production, coagulation, deposition, and transport because of ventilation [16]. With the support of the Nanotransport Project by the European Commission under FP6, Seipenbusch et al. performed a comparative study of the temporal evolution of platinum ENPs in a simulated chamber by using both experiment and simulation but failed to provide the comparative information about aggregate size-resolved distribution because they used a highly simplified coagulation model [8]. Anand et al. proceeded to perform PBM modeling with the Fuchs collision rate according to Seipenbusch et al. experiment [17]; they only reported a qualitative comparison rather than a quantitative comparison between PBM and the measurement for size-resolved PSD during evolution; thus, their study could not examine the extent to which PBM agrees with real physics. In their PBM experiment, the pre-exponential factor was specified as 1.0; many researchers have verified this value as unsuitable for the diffusion-limited aggregate process [9,10]. The study verified this claim (Section 3.6). Although Rim et al. [18]conducted a comparative study on the sized-resolved evolution of indoor ultrafine particles by using both experiments and PBM simulation, they emphasized the assessment of the relative relevance of different dynamical processes under indoor aerosol concentrations, and their collision kernel was limited to the modified Fuchs model. The improved collision rate, such as that reported in Thajudeen et al.'s model [9] designed for fractal-like aggregates, was not used; thus, the reliability of such collision rate models when applied in resolving ENP dynamics remains unclear. Studies have reported relevance of the size-resolved study of aerosols; however, no study has focused on the evaluation of the feasibility of PBM for ENPs under high concentration.

Considering ENPs in chemical reactors or workplaces, the correct acquisition of appropriate numbers and short sudden emission events are crucial. High emission with high particle emission rate is the key characteristic of such events [8]. This is in contrast to common studies on indoor aerosols, where the total particle number is not high [18–20]. Because of the high particle number, coagulation might play a more critical role in determining the rapid change in PSD than other dynamics; thus, while implementing PBM, a highly reliable collision rate model for aggregates rather than for spherical particles is essential. Since the pioneering work of Fuchs for spherical particle collision over the entire size regime