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Numerical study of nanoparticle formation in a free turbulent jet

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Abstract. Di-ethyl-hexyl-sebacate (DEHS) aerosol nanoparticle formation in a free turbulent jet as a result of nucleation, condensation and coagulation is studied using fluid flow simulation and the method of moments under the assumption of lognormal particle size distribution. The case of high nucleation rates and the coagulation-controlled growth of particles is considered. The formed aerosol performance is jet is numerically investigated for the various nozzle diameters and two approximations of the saturation pressure dependence on the temperature. It is demonstrated that a higher polydispersity of the aerosol is obtained for smaller nozzle diameters.

1. Introduction

Nanoparticle formation in a turbulent jet occurs in nature and industry for various practical cases. Aerosol nanoparticles are formed in engine exhaust plumes and propagated in the environment. The turbulent gas-vapour jets in aerosol reactors are used to generate the particles of submicron size for calibration systems. As a step of the creation of a reference submicron particle generator, the aerosol formation and growth in the continuously stirred tank reactor was theoretically and experimentally investigated by Koch et. al. [1]. The reactor provides nucleation of nanosize droplets and their growth by condensation and coagulation in the hot gas-vapour free turbulent jet. The influence of the saturation temperature and the flow rate on the particle size distribution (PSD) of the feed aerosol in the reactor was studied theoretically by Gilfanov et. al. [2]. The next important parameter that influences on the properties of aerosol is the jet nozzle diameter. The objective of this contribution is the study of the influence of the variation of the jet nozzle diameter on the PSD of the generated aerosol on the base of the early developed mathematical model.

2. Problem formulation

A hot gas-vapour turbulent jet that is a feed aerosol source in a continuously stirred tank reactor is considered (figure 1) [1, 2]. Di-ethyl-hexyl-sebacate (DEHS) vapour, saturated at temperature T_0, and nitrogen mixture streams with the velocity U_0 through a small nozzle of diameter d into the cubical mixing reactor with the side length of 60 cm. The gas temperature T_{amb} in the camera is assumed to be

much lower than the vapour saturation temperature. Due to mixing of hot jet with the surrounding cold gas the high supersaturation zone occurs at a distance of few diameters adjacent to the nozzle. As a result at high nucleation rates the formation of nanosize droplets takes place. Further growth of particles is performed due to condensation and coagulation in the jet mixing zone.

The gas phase is assumed to be a viscous compressible fluid. The steady turbulent fluid flow is described by the Reynolds-averaged Navier-Stokes equations. The realizable k-ε model is used to model turbulence. The temperature field is governed by the energy equation in the form

$$\nabla \cdot (\bar{u}(\rho E + p)) = \nabla \cdot ((k + \mu_t / \mathrm{Pr}_t)\nabla T), \tag{1}$$

where $\bar{u}$ is the averaged-gas velocity, ρ is the gas phase density, p is the pressure, $E = h - p/\rho + u^2/2$ is the specific energy, h is the sensible enthalpy, k is the thermal conductivity, μ_t is the turbulent viscosity, Pr_t is the turbulent Prandtl number. In the assumption of the axial symmetry the gas phase flow equations are solved in a cylindrical coordinate system. The ideal gas state law is used to close the system of fluid flow equations. The mathematical model of aerosol particle dynamics is based on the general dynamic equation (GDE) [3]

$$\begin{aligned}&\frac{\partial \rho n}{\partial t} + \nabla \cdot (\rho \bar{u} n) + \frac{\partial (G \rho n)}{\partial v} = \nabla \cdot (D \nabla n) + I(v^*)\delta(v - v^*) + \\ &+ \frac{1}{2}\int_0^v \beta(v - v', v')\rho n(v - v', t)\rho n(v', t)dv' - \rho n(v, t)\int_0^\infty \beta(v, v')\rho n(v', t)dv',\end{aligned} \tag{2}$$

where $n(v, t)$ is the PSD, $G(v)$ is the rate of condensational growth in the particle volume interval $[v, v+dv]$, D is the diffusivity of particles, I is the nucleation rate of new particles of critical volume v^*. The birth and loss of particles by coagulation in the size interval $[v, v+dv]$ are described by the last two terms in the RHS of Eq. (2), respectively. To solve Eq. (2) the method of moments is applied in the assumption of a lognormal shape of the PSD. This method was developed in previous works of Pratsinis [4], Lee [5, 6], Koch [7], Settumba and Garrick [8]. The transport equations for the moments M_k of the PSD obtained by multiplying Eq. (2) by v^k and integrating over the entire volume range are written in the form

$$\frac{\partial \rho M_k}{\partial t} + \nabla \cdot (\rho \bar{u} M_k) = \nabla \cdot (D \nabla M_k) + S_{M_k}, \tag{3}$$

where S_{Mk} is the source term accounting for internal processes: nucleation, condensation and coagulation. The total diffusivity is $D = D_m + D_t$, where D_m is the molecular diffusion coefficient and D_t is the turbulent diffusion coefficient ($D_t = \mu_t / \mathrm{Sc}_t$, μ_t is the turbulent viscosity, Sc_t is the turbulent Schmidt number). Finally the problem is reduced to the system of equations for the moments M_0, M_1 and M_2. The introduced moments M_0 and M_1 give the total number concentration of particles and the volume fraction of particles, respectively. To take into account gas phase-to-particle conversion the equation for the vapour concentration is solved together with Eq.(3).

The detailed description of the mathematical model of particle nucleation, condensation and coagulation for the considered problem was given in our previous paper [2]. The vapour saturation ratio, which is a ratio of the actual vapour pressure to the saturation pressure P_s, strongly affects the particle nucleation and condensation intensity. The dependence of saturation pressure on temperature can be found from experimental studies. The following two formulas were used to approximate $P_s(T)$ for the considered DEHS vapour

$$\log_{10} P_s = 15.3487 - 7230.347/(T + 51.865) \tag{4}$$

$$\log_{10} P_s = 10.371 - 3276.8155/(T - 91.4724) \tag{5}$$

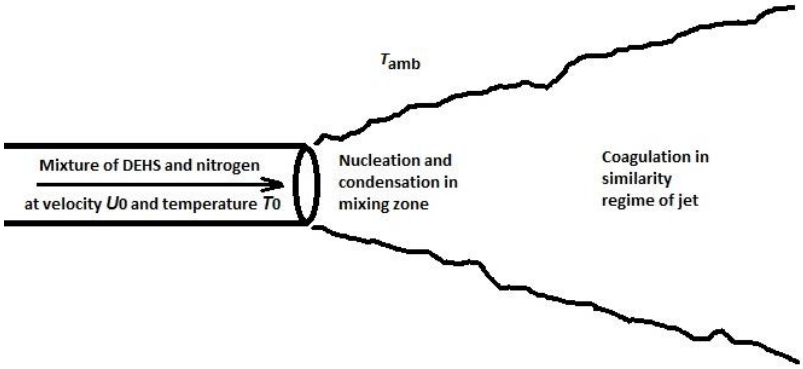

Figure 1. Flow diagram.

3. Results and discussion

To solve the fluid flow and aerosol dynamics equations the CFD package ANSYS Fluent was applied. For the accepted axial symmetry case the computational domain was a trapezoidal area with the length of 600*d* and heights of 80*d* and 120*d*. The structured grid composed by 601 and 401 points in axial and radial directions respectively is used for numerical calculations. The boundary conditions were the same as the conditions used in works [1, 2]: the exit velocity is $U_0 = 45$ m/s, the saturation temperature is $T_0 = 453$K, the ambient temperature is $T_{amb} = 295$ K. For the considered inlet diameters $d = 0.5$, 1 and 2 mm the Reynolds numbers are Re ≈ 700, 1400 and 2800, respectively. The turbulent Prandtl and Schmidt numbers were set to 0.85. The equations (3) for the moments were solved using User-Defined Scalars (UDS). The source terms in (3) were implemented by User-Defined Functions (UDF). The numerical scheme MUSCLE of third order accuracy was applied.

The lognormal PSD is determined by the geometric mean diameter d_g and the geometric standard deviation σ_g

$$n(v,t) = \frac{M_0}{\sqrt{2\pi} d_p \ln \sigma_g} \exp\left(-\frac{\ln^2(d/d_g)}{2\ln^2 \sigma_g} \right), \tag{6}$$

where d_p is the particle diameter. The particle number current $\dot{N}$ is calculated as

$$\dot{N}(x) = 2\pi \int_0^\infty r u_x(x,r) \rho M_0(x,r) dr, \tag{7}$$

where x and r are the axial and radial coordinates, respectively.

Using the described mathematical model the effect of various nozzles diameters was investigated numerically. The calculated axial gas phase velocity profiles are shown in figure 2a. The free jet with the smaller nozzle diameter is quenched faster that influences on the particle growth. The particle formation depends on the particle residence time τ_{res} that is defined as time for a particle to reach the current position on the axis. The residence time against the axial coordinate x for the various nozzle diameters is shown in figure 2b. The ratio of τ_{res} for the considered diameters is almost constant along the jet axis. The ratio of τ_{res} for $d = 0.5$ mm and 1 mm is about 2, for $d = 0.5$ mm and 2 mm it is 3.8, for $d = 1$ mm and 2 mm the ratio of τ_{res} is 1.9. This means that, for instance, for $d = 0.5$ mm the particles move to the final point 3.8 times slower than for $d = 2$ mm and internal physical processes occur 3.8 times longer.

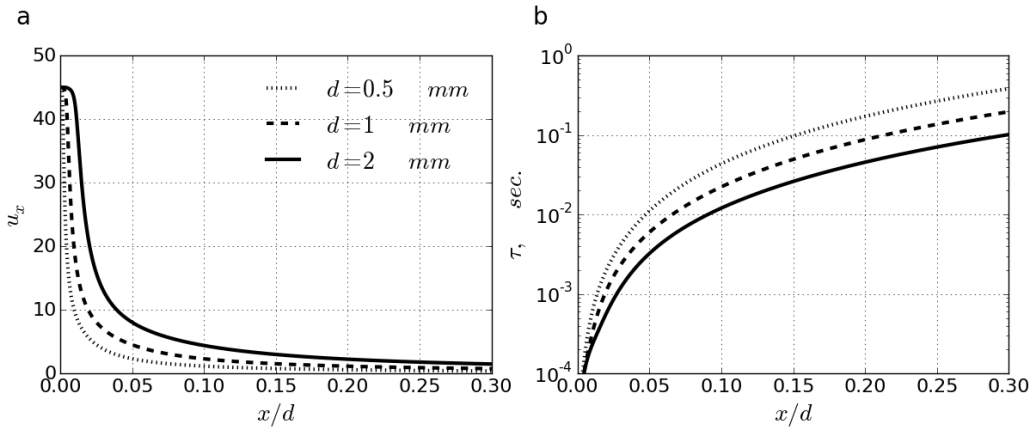

Figure 2. Profiles of axial velocity (a) and residence time of the particle (b) on the jet axis for different nozzle diameters.

The distributions of the particle number current $\dot{N}$, the geometric mean diameter d_g and the geometric standard deviation σ_g along the jet axis is shown in figure 3. The curves without and with markers are obtained using Eq. (4) and (5) respectively. Note that at T_0 = 453K the aerosol volume fractions of $1.3\cdot10^{-6}$ and $2.5\cdot10^{-6}$ are obtained for formulas (4) and (5), respectively. This leads to more notable production and growth effects of particles for the second approximation. As was mentioned the increase of nozzle diameter leads to the corresponding increase of flow rate. For a fixed value of the exit velocity (U_0 = 45 m/s), a twofold increase in the nozzle diameter leads to the same increase of Reynolds number, and a fourfold increase in flow rate. This results in the increase of the particle number current $\dot{N}$. However, the maximal values of $\dot{N}$ are sufficiently close to each other: $2.9\cdot10^{12}$-$4.1\cdot10^{12}$, $4.7\cdot10^{12}$-$6.4\cdot10^{12}$ and $7.2\cdot10^{12}$-$1.1\cdot10^{13}$ s^{-1}. The two numbers in the given ranges here and further correspond to the limits found using Eq. (4) and Eq. (5). The initial growth of particles by condensation has the larger contribution for larger diameters. For example at a distance of 20*d* from the nozzle the values d_g are 19-22, 26-30 and 36-40 nm for d = 0.5, 1 and 2 mm, respectively. It is seen from figure 3b that the growth of particles by coagulation is more considerable for smaller nozzle diameters. The number current decreases 4.9-6.6 times (to $5.9\cdot10^{11}$-$6.2\cdot10^{11}$ s^{-1}), 2.5-3 times (to $1.9\cdot10^{12}$-$2.1\cdot10^{12}$ s^{-1}) and 1.5-1.8 times (to $4.7\cdot10^{12}$-$6.0\cdot10^{12}$ s^{-1}), the geometric mean diameter increases 1.57-1.68 times (to 30-37 nm), 1.27-1.33 times (to 33-40 nm) and 1.1-1.15 times (to 40-46 nm) for d = 0.5, 1 and 2 mm, respectively. As it can be expected all the effects (nucleation, condensation and coagulation) are more essential for Eq. (5) as in this case the mass fraction of aerosol is two times larger. The geometric standard deviation σ_g increases quickly to 1.3 for the smallest diameter (especially for Eq. (5)) and sufficiently slow to 1.23 for largest one. The values of $\sigma_g > 1.25$ and $\sigma_g < 1.25$ for d = 0.5 mm and d = 2 allow us to conclude that we have polydisperse and monodisperse aerosol formation in these two cases. The effect can be explained by the fact that for the smaller diameter locally the higher particle number concentration is produced by nucleation. This is shown in figure 4a where axial profiles of the first moment M_0 are plotted. The maximum of the number concentration is about 2 and 4 times larger for d = 0.5 m than for d = 1 mm and d = 2 mm respectively. The collision time τ_{coll} is a characteristic time of coagulation process and is inversely related with particle concentration M_0. We can see in figure 4b that the collision time for the smallest diameter is lower at the initial stage of the free jet and coagulation is much more intensive. However, at the late stages of the free jet the pattern is reversed. The higher particle concentration with the higher residence time makes the particle formation for smaller d more controlled by coagulation. It was shown in [2] that the coagulation-controlled particle growth regime is more stable relatively to the change of external conditions. So in the context of using free turbulent jet as a feeding system for the reference particle size distribution generator smaller nozzle diameters can be recommended. Another effect in behavior of collision time for different saturation pressure curves can be seen in figure 4b.

The collision time obtained using Eq. (5) is less than one obtained using Eq. (4) at the beginning of the jet as much more particles are produced in this case (figure 4a). However particles are spent to coagulation more intensively for Eq. (5) and the collision time grows faster along jet axis.

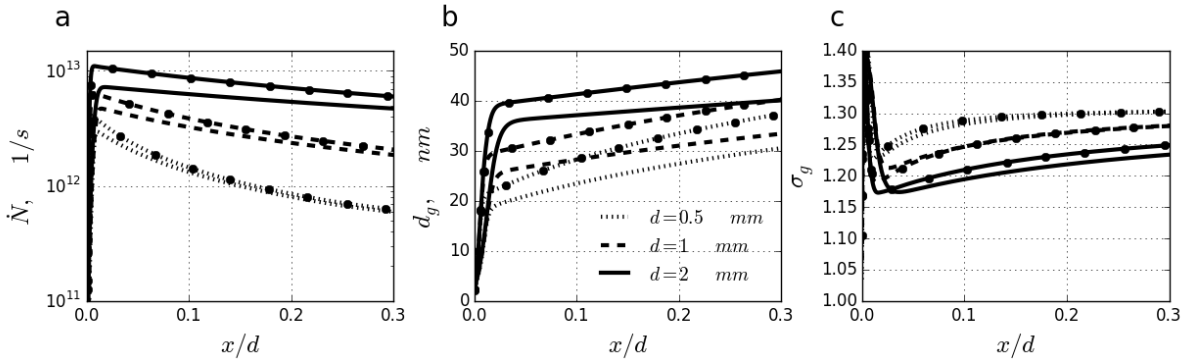

Figure 3. Profiles of particle number current (a), geometric mean diameter (b) and standard geometric deviation (c) on the jet axis for different nozzle diameters.

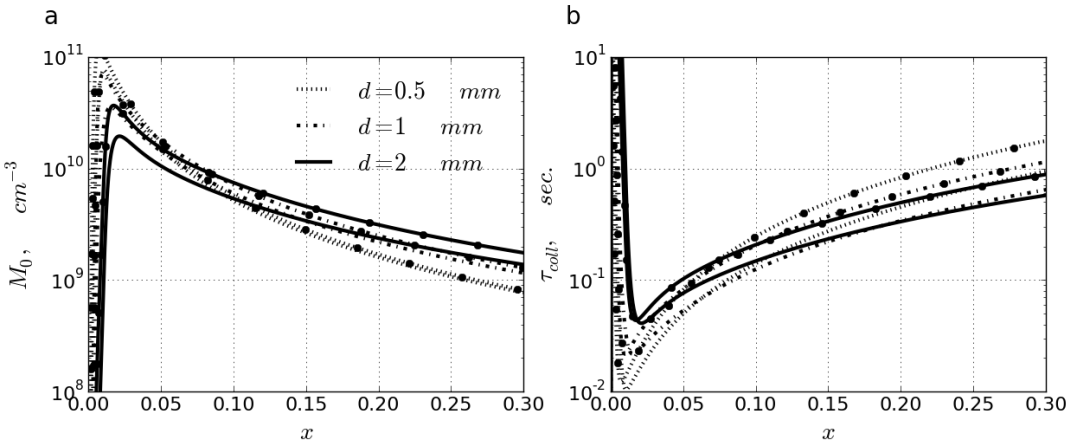

Figure 4. Profiles of particle concentration (a) and collision time (b) on the jet axis for different nozzle diameters.

4. Conclusions

The mathematical model of aerosol particle formation in a hot turbulent gas-vapour jet is used to simulate the nanoparticles generation as a result of nucleation, condensation and coagulation processes. The model includes the Reynolds-averaged Navier-Stokes equations for the fluid flow and general aerosol dynamics equation. The moment method for the lognormal particle size distribution is applied. The influence of the variation of the nozzle diameter on the generated aerosol characteristics is investigated. As a result of the parametrical calculations it is found that for the larger gas-vapour nozzle diameters an aerosol with higher particle number current and mean particle size is obtained. The use of the smaller nozzle diameters leads to higher particle concentration and aerosol growth in the coagulation-controlled mode that allows obtaining the aerosol with higher polydispersity. It means that the smaller nozzle diameters can be recommended for the design of the reference particle size distribution generator to realize more stable aerosol formation.

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References

[1] Koch W, Lödding H and Pohlmann G. 2012 *J. Aerosol Sci* **49** 1-8.
[2] Gilfanov A K, Koch W and Zaripov S K 2016 *J. Aerosol Sci* **96** 124-139.
[3] Friedlander S K 2000 *Smoke, dust, and haze. Fundamentals of aerosol dynamics* (New York: Oxford University Press).
[4] Pratsinis S E 1988 *J Colloid Interf Sci* **124** 416-427.
[5] Lee K W and Chen H 1984 *Aerosol Sci Tech* **3** 327-334.
[6] Lee K W, Chen H and Gieseke J A 1984 *J. Aerosol Sci* **3** 53-62.
[7] Koch W, Windt H and Karfich N 1993 *J. Aerosol Sci* **28** 909-918.
[8] Settumba N and Garrick S C 2003 *J. Aerosol Sci* **34** 149-167.