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# Simulation of nanoparticle synthesis in an aerosol flame reactor using a coupled flame dynamics–monodisperse population balance model

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ABSTRACT

Flame aerosol synthesis is one of the commonly employed techniques for producing ultra fine particles of commodity chemicals such as titanium dioxide, silicon dioxide and carbon black. Large volumes of these materials are produced in industrial flame reactors. Particle size distribution of product powder is the most important variable and it depends strongly on flame dynamics inside the reactor, which in turn is a function of input process variables such as reactant flow rate and concentration, flow rates of air, fuel and the carrier gas and the burner geometry. A coupled flame dynamics–monodisperse population balance model for nanoparticle synthesis in an aerosol flame reactor is presented here. The flame dynamics was simulated using the commercial computational fluid dynamics software CFX and the particle population dynamics was represented using a monodisperse population balance model for continuous processes that predicts the evolution of particle number concentration, particle volume and surface area. The model was tested with published experimental data for synthesis of silica nanoparticles using different burner configurations and with different reactor operating conditions. The model predictions for radial flame temperature profiles and for the effects of process variables like precursor concentration and oxygen flow rate on particle specific surface area and mean diameter are in close agreement with published experimental data.



## 1. Introduction

Many commodity and specialty chemicals and materials such as carbon black, titania, silica and zinc oxide are produced in the form of fine particles, which find applications in a wide variety of industrial and domestic products ranging from tires, printing inks, paints and pigments, plastics, optical fibers, catalysts, pharmaceutical powders and cosmetics. For many practical applications, it is desirable to have particles of small size and a narrow size distribution because smaller particles with narrow size distribution result in better properties of finished products. For example, activity of catalysts, hardness and strength of metals and electrical conductivity of ceramics improve as the particle size decreases (Gutsch et al., 2002). Flame aerosol synthesis is one of the commonly employed techniques for producing fine particles on the industrial scale. This process has a significant advantage over its rivals, in that, it offers many control variables like flame temperature, flame structure, stoichiometry, pressure level, residence time distribution, turbulence, etc. (Rosner, 2005).

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## Nomenclature

| | |
|---|---|
| $a_p$ | monomer surface area, $m^2$ |
| $A$ | total particle surface area density, $m^2\ m^{-3}$ |
| $A_{EDM}$ | constant in the Eddy-dissipation model |
| $A_{min}$ | minimal total surface area density, $m^2\ m^{-3}$ |
| $B_{EDM}$ | constant in the Eddy-dissipation model |
| $c$ | particle velocity, $m\ s^{-1}$ |
| $c_i$ | concentration of component $i$ in the gas, $kmol\ m^{-3}$ |
| $C_{O_2}$ | concentration of oxygen, $kmol\ m^{-3}$ |
| $C_{precursor}$ | concentration of the precursor, $kmol\ m^{-3}$ |
| $d_n$ | number-averaged particle diameter, m |
| $d_p$ | primary particle diameter, m |
| $D$ | particle diffusion coefficient, $m^2\ s^{-1}$ |
| $D_{SiO_2-O_2}$ | diffusion coefficient of silica through $O_2$, $m^2\ s^{-1}$ |
| $E$ | activation energy, $J\ kmol^{-1}$ |
| $\mathbf{g}$ | gravitational acceleration, $m\ s^{-2}$ |
| $G$ | total growth rate of a particle, $m\ s^{-1}$ |
| $G_M$ | growth rate of a particle under the control of mass transfer, $m\ s^{-1}$ |
| $G_C$ | growth rate of a particle under the control of chemical reaction kinetics, $m\ s^{-1}$ |
| $h$ | specific enthalpy of the gas, $J\ kg^{-1}$ |
| $I_N$ | rate of nucleation, $m^{-3}\ s^{-1}$ |
| $\mathbf{J}_i$ | diffusion flux of species $i$, $kg\ m^{-2}\ s^{-1}$ |
| $k$ | turbulent kinetic energy, $m^2\ s^{-2}$ |
| $k_0$ | pre-exponential factor in the Arrhenius rate expression for the precursor oxidation, $m^3\ kmol^{-1}\ s^{-1}$ |
| $k_g$ | thermal conductivity of the gas, $J\ m^{-1}\ s^{-1}\ K^{-1}$ |
| $k_{gc}$ | particle growth rate constant, $m\ s^{-1}$ |
| $k_n$ | nucleation rate constant, $m^{-3}\ s^{-1}$ |
| $Kn$ | Knudsen number |
| $N$ | particle number concentration, $m^{-3}$ |
| $N_g$ | parameter in the expression for the particle growth rate controlled by chemical reaction |
| $N_n$ | exponent in the equation for rate of nucleation |
| $p$ | pressure, Pa |
| $P_k$ | turbulence production due to viscous and buoyancy forces, $kg\ s^{-3}$ |
| $P_{kb}$ | turbulence production due to buoyancy forces, $kg\ s^{-3}$ |
| $r_c$ | particle collision radius, m |
| $R$ | gas constant, $J\ kmol^{-1}\ K^{-1}$ |
| $R_1$ | rate of mixing of the reactants in the turbulent eddies, $kmol\ m^{-3}\ s^{-1}$ |
| $R_2$ | rate of mixing of the hot product gases with the cold reactant gases, $kmol\ m^{-3}\ s^{-1}$ |
| $R_i$ | net rate of production of species $i$ by a chemical reaction, $kg\ m^{-3}\ s^{-1}$ |
| $R_A$ | rate of oxidation of the precursor using the Arrhenius expression, $kmol\ m^{-3}\ s^{-1}$ |
| $S$ | degree of supersaturation |
| $S_E$ | heat source term that denotes the rate of heat liberated due to chemical reaction and heat absorption due to radiation, $J\ m^{-3}\ s^{-1}$ |
| $S_i$ | rate of creation of species $i$ by addition from the dispersed phase, $kg\ m^{-3}\ s^{-1}$ |
| $S_V$ | rate of increase of total particle mass per unit volume of the system, $kg\ m^{-3}\ s^{-1}$ |
| $t_p$ | transition parameter in the expression for coagulation rate coefficient, $\beta$ |
| $T$ | temperature, K |
| $\mathbf{u}$ | gas velocity, $m\ s^{-1}$ |
| $v_p$ | monomer volume, $m^3$ |
| $V$ | total particle volume density, $m^3\ m^{-3}$ |
| $V_{mol}$ | molar volume, $m^3\ mol^{-1}$ |
| $W_i$ | molar mass of the component $i$, $kg\ kgmol^{-1}$ |
| $Y^*_{SiO_2}$ | mole fraction of species 'A' at saturation temperature $T$ |
| $Y_i$ | mass fraction of component $i$ in the gas |

*Greek letters*

| | |
|---|---|
| $\beta$ | coagulation rate coefficient, $m^3\ s^{-1}$ |
| $\varepsilon$ | dissipation rate of turbulent kinetic energy, $m^2\ s^{-3}$ |
| $\lambda$ | gas mean free path, m |
| $\mu$ | gas viscosity, $kg\ m^{-1}\ s^{-1}$ |
| $\mu_t$ | turbulent gas viscosity, $kg\ m^{-1}\ s^{-1}$ |
| $v_i$ | stoichiometric coefficient of component $i$ |
| $\rho$ | gas density, $kg\ m^{-3}$ |
| $\boldsymbol{\tau}$ | stress tensor, $kg\ s^{-2}\ m^{-1}$ |
| $\boldsymbol{\tau}^{(t)}$ | turbulent momentum flux tensor, $kg\ s^{-2}\ m^{-1}$ |
| $\tau_f$ | characteristic coalescence time, s |

But the powders produced in the aerosol flame reactors have a relatively large particle size and have wide size distribution, with particle size ranging from a few nanometers to several micrometers (Ulrich, 1984). There is a need for controlling the product particle size distribution by manipulating process input variables. On the other hand, nanoparticles of a variety of materials have been produced on the laboratory scale in aerosol flame reactors but scaling up the process to produce nanoparticles in large quantities, while maintaining the particle size in the nanometer range, still remains a challenging task.

Nanoparticles are synthesized using either the premixed or the diffusion flame burners on the laboratory scale. The precursor or reactant is injected into the reactor in the form of a vapor or liquid using a carrier gas along with air and fuel. Chemical reactions occur in the gas phase in the flame at high temperatures and result in product molecules, which grow further by coagulation and/or surface reaction and form product particles. Later on, as the temperature decreases, particle growth takes place mainly by coagulation and results in irregularly structured particles. The product particle size distribution is influenced by several factors, namely, burner geometry, nature of flame and its configuration, inlet reactant