Nonlinear control of spatially inhomogenous aerosol processes

Ashish Kalani, Panagiotis D. Christofides*

Department of Chemical Engineering, School of Engineering and Applied Sciences, University of California, Los Angeles, CA 90095-1592, USA

Abstract

This article proposes a nonlinear feedback control methodology for spatially inhomogeneous aerosol processes for which the manipulated inputs, the control objectives and the measurements are distributed in space. Initially, a general nonlinear partial integro-differential equation model which describes the spatio-temporal evolution of the aerosol size distribution, as well as the evolution of the concentrations of species and temperature of the continuous phase is presented. The model accounts for simultaneous chemical reaction, nucleation, condensation, coagulation and convective transport. Then, under the assumption of lognormal aerosol size distribution, the method of moments is employed to reduce the original model into a set of first-order hyperbolic partial differential equations (PDEs) which accurately describes the spatio-temporal evolution of the three leading moments needed to precisely characterize the aerosol size distribution. This hyperbolic PDE system is then used as the basis for the synthesis of nonlinear distributed output feedback controllers that enforce closed-loop stability and achieve an aerosol size distribution with desired characteristics. The proposed nonlinear control method is successfully applied through simulations to a typical aerosol process and is shown to outperform a conventional proportional integral control scheme and deal effectively with disturbances in the feed to the process. © 1999 Elsevier Science Ltd. All rights reserved.

1. Introduction

Aerosol processes are widely used in industry for the production of fine particles including pigments, carbon black, optical fibers, silicon and ceramic powders, and are characterized by coupled chemical reaction, nucleation, condensation and coagulation phenomena. In particular, in a typical aerosol process, aerosol precursors (molecules of gaseous reactants) react to produce a monomer (molecule of condensable species). The monomer molecules form unstable clusters, which grow further by monomer condensation. Beyond a critical cluster size, nucleation of aerosol particles occurs. These particles grow further, mainly by coagulation.

The dynamic models of aerosol processes are typically obtained from the application of population, material and energy balances, and consist of nonlinear partial integro-differential equation systems (where the independent variables are time, space and one or more internal particle coordinates, such as particle volume, shape). Nonlinearities usually arise from complex reaction, nu-

cleation, condensation and coagulation rates and their nonlinear dependence on temperature. The complex nature of aerosol process models has motivated an extensive research activity on the development of numerical methods for the accurate computation of their solution. Examples of solution methods include the method of self-preserving distributions (e.g. Ramabhadran and Seinfield, 1975; Friedlander, 1977), the method of weighted residuals (Ramakrishna, 1985; Gelbard and Seinfield, 1978), the sectional method (e.g. Gelbard et al., 1980; Landgrebe and Pratsinis, 1990), and discretization via fixed/moving pivot techniques (e.g. Kumar and Ramakrishna, 1996a, b). However, even though the above methods can produce accurate solutions of aerosol process models, they are not suitable for the derivation of approximate models which can be used for the synthesis of practically implementable feedback controllers. Fortunately, the aerosol size distributions obtained by many industrial reactors can be adequately described by lognormal functions, and thus, the dominant dynamic behavior of many aerosol processes can be accurately captured by a model that describes the evolution of the leading moments of the aerosol size distribution (Pratsinis, 1988; Williams and Loyalka, 1991). Moment models are much simpler to analyze, simulate and utilize for

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*Corresponding author. Tel.: 001 310 794 1015; fax: 001 310 206 4107.

E-mail address: pdc@seas.ucla.edu. (P.D. Christofides)

controller design than detailed aerosol process models, while they contain the most important information about the dynamics of the aerosol process.

In spite of the vast amount of literature on solution methods and dynamical analysis for aerosol processes, few papers are available on feedback control of aerosol processes based on nonlinear population balance models. In fact, even the broader subject of control of particulate processes based on population balance models has received very little attention. More specifically, previous research on control of particulate processes has mainly focused on: (a) the understanding of controllability and observability properties of population balance models (Heshemi and Epstein, 1982; Semino and Ray, 1995a), (b) stability analysis using Laplace transform and Lyapunov functionals (e.g. Hale and Verduyn Lunel, 1993; Rawlings and Ray, 1987), and (c) the application of conventional control schemes (e.g. proportional-integral, proportional-integral-derivative, self-tuning control) to crystallizers and emulsion polymerization processes (e.g. Semino and Ray, 1995b; Rohani and Bourne, 1990; Dimitratos et al., 1994). These conventional control schemes clearly limit the achievable control quality and may lead to poor performance, or even to destabilization of the closed-loop system, in the presence of severe process nonlinearities. A notable exception on model-based control of particulate processes is an optimization-based control method which was developed in Eaton and Rawlings (1990) (see also Rawlings et al., 1993) and successfully applied to a batch crystallization process.

In this work, we develop a nonlinear feedback control method for spatially inhomogeneous aerosol processes for which the manipulated inputs, the control objectives and the measurements are distributed in space. Initially, a general nonlinear partial integro-differential equation model which describes aerosol processes with simultaneous chemical reaction, nucleation, condensation, coagulation and convective transport is presented. Then, under the assumption of lognormal aerosol size distribution, the method of moments is employed to reduce the original model into a set of first-order hyperbolic PDEs which accurately describes the spatio-temporal evolution of the three leading moments needed to exactly characterize the aerosol size distribution. This hyperbolic partial differential equations (PDEs) system is then used as the basis for the synthesis of nonlinear distributed output feedback controllers that use process measurements to achieve an aerosol size distribution with desired characteristics (e.g. geometric average particle volume). The controller design problem is addressed by using recent results on nonlinear control of hyperbolic PDE systems (Christofides and Daoutidis, 1996). The performance of the proposed control method is successfully tested through simulations on a typical aerosol process and is shown to be superior to the one of a proportional integral controller.

2. Preliminaries

2.1. Spatially inhomogeneous aerosol process model

We consider aerosol processes with simultaneous chemical reaction, nucleation, condensation, coagulation and convective transport. Fig. 1 shows one such aerosol process in a typical aerosol flow reactor. A general mathematical model which describes the spatio-temporal evolution of the particle size distribution in such aerosol processes can be obtained from a population balance and consists of the following nonlinear partial integro-differential equation:

$$\frac{\partial n}{\partial t} + v_z \frac{\partial n}{\partial z} + \frac{\partial (G(\bar{x}, v, z)n)}{\partial v} - I(v^*)\delta(v - v^*)$$

$$= \frac{1}{2} \int_0^v \beta(v - \bar{v}, \bar{v}, \bar{x})n(v - \bar{v}, z, t)n(\bar{v}, z, t)\,d\bar{v}$$

$$- n(v, z, t) \int_0^\infty \beta(v, \bar{v}, \bar{x})n(\bar{v}, z, t)\,d\bar{v}, \tag{1}$$

where $n(v, z, t)$ denotes the particle size distribution function, v is the particle volume, t is the time, $z \in [0, L]$ is the spatial coordinate, L is the length of the process, v_z is the velocity of the fluid, $G(\bar{x}, v, z)$, $I(v^*)$, $\beta(v - \bar{v}, \bar{v}, \bar{x})$ are nonlinear scalar functions and $\delta(\cdot)$ is the standard Dirac function.

On the other hand, a mathematical model which predicts the spatio-temporal evolution of the concentrations of species and temperature of the gas phase can be obtained from mass and energy balances and has the following form:

$$\frac{\partial \bar{x}}{\partial t} = \bar{A} \frac{\partial \bar{x}}{\partial z} + \bar{f}(\bar{x}) + \bar{g}(\bar{x})\,b(z)\,u(t) + \tilde{A} \int_0^\infty a(\eta, v, x)\,dv, \tag{2}$$

where $\bar{x}(z, t)$ is an n-dimensional vector of state variables that depend on space and time, $\bar{A}, \tilde{A}$ are constant matrices, $\bar{f}(\bar{x}), \bar{g}(\bar{x}), a(\eta, v, x)$ are nonlinear vector functions, $u(t)$ is the axially distributed manipulated input (e.g. wall temperature) and $b(z)$ is a known function which determines how the control action $u(t)$, is distributed in space
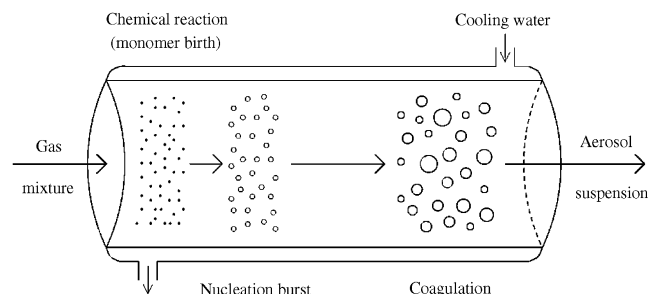

Fig. 1. A typical aerosol flow reactor.

(for example, $b(z) = 1$ when $u(t)$ is uniformly distributed in space). The term $\tilde{A} \int_0^\infty a(\eta, v, x) dv$ accounts for mass and heat transfer from the continuous phase to all the particles in the population.

In the population balance of Eq. (1), the term $\partial n / \partial t$ describes the rate of change of particle concentration in the particle volume interval v, $v + dv$ and in the spatial interval z, $z + dz$, and the term $v_z \partial n / \partial z$ corresponds to convective transport of aerosol particles at velocity v_z. Moreover, the terms $\partial(G(\bar{x}, v, z)n)/\partial v$ and $I(v^*)\delta(v - v^*)$ account for the loss or gain of particles by condensation at rate G and the formation of new particles of critical volume v^* by nucleation at rate I, respectively. Finally, the gain and loss of particles by Brownian coagulation is captured by the terms

$$\frac{1}{2} \int_0^v \beta(v - \bar{v}, \bar{v}, \bar{x})n(v - \bar{v}, z, t)n(\bar{v}, z, t) d\bar{v}, \quad \text{and}$$

$$n(v, z, t) \int_0^\infty \beta(v, \bar{v}, \bar{x})n(\bar{v}, z, t) d\bar{v}, \tag{3}$$

respectively. $G(\bar{x}, v, z)$ and β are the condensational growth and collision frequency function, respectively, for which two different expressions are used for the free molecule size and continuum size regimes (Pratsinis, 1988): (a) for the free molecule size regime:

$$G_{FM}(\bar{x}, v, z) = B_1 v^{1/3}(S - 1),$$

$$B_1 = (36\pi)^{1/3} v_1 n_s (k_B T / 2\pi m_1)^{1/2}$$

$$\beta_{FM}(\bar{x}, v, z) = B_2 \left(\frac{1}{v} + \frac{1}{\bar{v}}\right)^{1/2} (v^{1/3} + \bar{v}^{1/3})^2,$$

$$B_2 = (3/4\pi)^{1/6} (6k_B T v_1 / m_1)^{1/2}, \tag{4}$$

and (b) for the continuum size regime:

$$G_C(\bar{x}, v, z) = B_3 v^{1/3}(S - 1),$$

$$B_3 = (48\pi^2)^{1/3} D v_1 n_s,$$

$$D = \lambda (8 k_B T / \pi m_1)^{1/2} / 3,$$

$$\beta_C = B_4 \left(\frac{C(v)}{v^{1/3}} + \frac{C(\bar{v})}{\bar{v}^{1/3}}\right)(v^{1/3} + \bar{v}^{1/3}),$$

$$B_4 = \frac{2k_B T}{3\mu}. \tag{5}$$

In Eqs. (4) and (5), S is the saturation ratio, T is the temperature, D is the condensable vapor diffusivity, λ is the mean free path of the gas, μ is the viscosity of the fluid, n_s is the monomer concentration at saturation ($n_s = P_s / k_B T$, where P_s is the saturation pressure), m_1 is the monomer mass, v_1 is the monomer volume, r is the particle radius, $C(v) = 1 + B_5 \lambda / r$ is the Cunningham correction factor and $B_5 = 1.257$. Finally, the nucleation rate $I(v^*)$ is assumed to follow the classical Becker–Dor-

ing theory and is given by the following expression (Pratsinis, 1988):

$$I = n_s^2 s_1 (k_B T / 2\pi m_1)^{1/2} S^2 (2/9\pi)^{1/3} \Sigma^{1/2} \exp(-k^* \ln S / 2), \tag{6}$$

where s_1 is the monomer surface area and k^* is the number of monomers in the critical size nucleus which is given by:

$$k^* = \frac{\pi}{6} \left(\frac{4\Sigma}{\ln S}\right)^3, \tag{7}$$

where $\Sigma = \gamma v_1^{2/3} / k_B T$ and γ is the surface tension.

Remark 1. Referring to the system of Eqs. (1) and (2), several remarks are in order: (a) the spatial differential operators are linear; this assumption is valid for spatially inhomogeneous aerosol processes for which the particle and gas velocities can be considered to be independent of temperature, concentrations and particle size distribution; (b) plug-flow is considered and diffusive phenomena in radial and axial directions are neglected; these assumptions are made to simplify the development; (c) turbulent coagulation is neglected; this assumption is made to simplify the notation of the theoretical results of the paper and can be readily relaxed; (d) the effect of particle curvature on aerosol evaporation rate (Kelvin effect) is neglected; this is done to achieve closure of the integral (moment) aerosol equations (see Section 3 below); (e) the manipulated input $u(t)$, is a lumped variable (i.e., independent of the spatial coordinate z) and enters the system through Eq. (2) (mass and energy balance model); this assumption is usually satisfied in most practical applications where the wall temperature is chosen as the manipulated input; and (f) the nonlinearities in Eq. (2) appear in an additive fashion (e.g. complex reaction rates, Arrhenius dependence of reaction rates on temperature).

2.2. Methodological framework for control of aerosol processes

The complexity of the partial integro-differential equation model of Eqs. (1) and (2) does not allow its direct use for the synthesis of a practically implementable nonlinear model-based feedback controller for spatially inhomogeneous aerosol processes. This obstacle, together with the experimental observation that many aerosol size distributions can be adequately approximated by lognormal functions, motivate employing the following methodology for controller design:

1. Initially, the aerosol size distribution is assumed to be described by a lognormal function and the method of moments is applied to the population balance of Eq. (1) to compute a hyperbolic PDE system (where the independent variables are time and space) that

describes the spatio-temporal behavior of the three leading moments needed to exactly describe the evolution of the lognormal aerosol size distribution.

2. Then, nonlinear geometric control methods (Christofides and Daoutidis, 1996) are applied to the resulting system of hyperbolic PDEs to synthesize nonlinear distributed output feedback controllers that use process measurements at different locations along the length of the process to adjust the manipulated input (typically, wall temperature), in order to achieve an aerosol size distribution with desired characteristics (e.g. geometric average particle volume).

3. Lognormal aerosol size distribution – moment model

In this section, we assume that the aerosol size distribution can be adequately represented by a lognormal function and use the method of moments to derive a PDE system which describes the spatio-temporal evolution of the three leading moments of the distribution. The assumption of lognormal aerosol size distribution is motivated by experimental results, as well as from the fact that the parameters of a lognormal function are very frequently used in practice to measure and characterize aerosol size distributions. More specifically, a lognormal aerosol size distribution is described by (Pratsinis, 1988)

$$n(v, z, t) = \frac{1}{3\sqrt{2\pi}\ln\sigma} \exp\left(-\frac{\ln^2(v/v_g)}{18\ln^2\sigma}\right)\frac{1}{v}, \tag{8}$$

where v_g is the geometric average particle volume and σ is the standard deviation. Defining the kth moment of the distribution as

$$M_k(t) = \int_0^\infty v^k n(v, z, t)\,dv, \tag{9}$$

one can exactly express v_g and σ in terms of the first three moments of the distribution according to the following relations (Brock et al., 1986):

$$v_g = \frac{M_1^2}{M_0^{3/2}M_2^{1/2}}, \qquad \ln^2\sigma = \frac{1}{9}\ln\left(\frac{M_0M_2}{M_1^2}\right). \tag{10}$$

Clearly, the precise characterization of a lognormal aerosol size distribution requires only knowledge of the three leading moments. Therefore, in the remainder of this section, we derive the PDE system that describes the evolution of the first three leading moments of the entire particle size distribution. To this end, we initially compute the moment models for the free molecule size regime and continuum size regime. These models are then used to derive the moment model that describes aerosol dynamics over the entire particle size spectrum by using the harmonic average of the condensation and coagulation rates in the free molecule and continuum regimes.

Free molecule size regime. The PDE system that describes the spatio-temporal evolution of the kth moment of the aerosol size distribution will be computed by substituting Eq. (4) into Eq. (1), multiplying by v^k, and integrating over all particle sizes. Following this procedure, one can show (the detailed calculations are omitted due to space limitations) that the evolution of M_0 (particle concentration), which is only affected by nucleation and coagulation, is governed by

$$\frac{\partial M_0}{\partial t} = -v_z\frac{\partial M_0}{\partial z} + I - b_0 B_2(M_{2/3}M_{-1/2}$$
$$+ 2M_{1/3}M_{-1/6} + M_{1/6}M_0), \tag{11}$$

where the coefficient b_0 is used for the relationship:

$$\left(\frac{1}{v} + \frac{1}{\bar{v}}\right)^{1/2} = b_0\left(\frac{1}{v^{1/2}} + \frac{1}{\bar{v}^{1/2}}\right) \tag{12}$$

and was computed by the expression $b_0 = 0.633 + 0.092\sigma^2 - 0.022\sigma^3$ (Pratsinis, 1988). The evolution of M_1 (aerosol volume), which is affected by nucleation and condensation, is given by

$$\frac{\partial M_1}{\partial t} = -v_z\frac{\partial M_1}{\partial z} + Iv^* + B_1(S-1)M_{2/3}. \tag{13}$$

Finally, the second moment, M_2, depends on nucleation, condensation and coagulation and is described by

$$\frac{\partial M_2}{\partial t} = -v_z\frac{\partial M_2}{\partial z} + Iv^{*2} + 2B_1(S-1)M_{5/3}$$
$$+ 2b_2 B_2(M_{5/3}M_{1/2} + 2M_{4/3}M_{5/6} + M_{7/6}M_1), \tag{14}$$

where b_2 is used as b_0 but for the coagulation kernel of the second moment and is computed by the expression $b_2 = 0.39 + 0.5\sigma - 0.214\sigma^2 + 0.029\sigma^3$ (Pratsinis, 1988).

Continuum size regime. Similar to the case of the free molecule regime, the spatio-temporal evolution of the kth moment of the aerosol size distribution in the continuum size regime is described by a PDE system, which can be obtained by substituting Eq. (5) into Eq. (1), multiplying by v^k, and integrating over all particle sizes. More specifically, the evolution of the zeroth, first and second moments is governed by the following PDEs:

$$\frac{\partial M_0}{\partial t} = -v_z\frac{\partial M_0}{\partial z} + I - B_4[M_0^2 + M_{1/3}M_{-1/3}$$
$$+ B_5\lambda(4\pi/3)^{1/3}(M_0M_{-1/3} + M_{1/3}M_{-2/3})], \tag{15}$$

$$\frac{\partial M_1}{\partial t} = -v_z\frac{\partial M_1}{\partial z} + Iv^* + B_3(S-1)M_{1/3}, \tag{16}$$

Table 1
Dimensionless variables

$N = M_0/n_s$	Aerosol concentration
$V = M_1/n_s v_1$	Aerosol volume
$V_2 = M_2/n_s v_1^2$	Second aerosol moment
$\tau = (2\pi m_1/k_B T)^{1/2}/n_s s_1$	Characteristic time for particle growth
$K = (2k_B T/3\mu)\, n_s \tau$	Coagulation coefficient
$K_{n_1} = \lambda/r_1$	Monomer Knudsen number
$I' = I/(n_s/\tau)$	Nucleation rate
$R'_r = R_r/(n_s/\tau)$	Reaction rate group
$v'_g = v_g/v_1$	
$r'_g = r_g/r_1$	
$\bar{z} = z/L$	
$v_{zl} = \tau v_z/L$	
$\theta = t/\tau$	

$$\frac{\partial M_2}{\partial t} = - v_z \frac{\partial M_2}{\partial z} + I v^{*2}$$
$$+ 2B_3(S-1)M_{4/3} + 2B_4[M_1^2 + M_{4/3}M_{2/3}$$
$$+ B_5\lambda(4\pi/3)^{1/3}(M_1 M_{2/3} + M_{1/3}M_{4/3})]. \tag{17}$$

Combining the moment models of Eqs. (11), (13) and (14) and Eqs. (15)–(17) by using the harmonic average of the condensation and coagulation rates in the free molecule and continuum regimes, we derive the following dimensionless (see Table 1 for the list of dimensionless variables) model for aerosol nucleation, condensation, coagulation and convective transport in the entire particle size spectrum.

Zeroth moment (*aerosol concentration*):

$$\frac{\partial N}{\partial \theta} = - v_{zl}\frac{\partial N}{\partial \bar{z}} + I' - \xi N^2. \tag{18}$$

where

$$\frac{1}{\xi} = \frac{1}{\xi_{FM}} + \frac{1}{\xi_C},$$

$$\xi_{FM} = r'^{1/2}_g b_0[\exp(\tfrac{25}{8}\ln^2\sigma) + 2\exp(\tfrac{5}{8}\ln^2\sigma)$$
$$+ \exp(\tfrac{1}{8}\ln^2\sigma)],$$

$$\xi_C = K[1 + \exp(\ln^2\sigma) + B_5(K_{n_1}/r'_g)$$
$$\times \exp(\tfrac{1}{2}\ln^2\sigma)(1 + \exp(2\ln^2\sigma))]. \tag{19}$$

First moment (*aerosol volume*):

$$\frac{\partial V}{\partial \theta} = - v_{zl}\frac{\partial V}{\partial \bar{z}} + I'k^* + \eta(S-1)N, \tag{20}$$

where

$$\frac{1}{\eta} = \frac{1}{\eta_{FM}} + \frac{1}{\eta_C},$$

$$\eta_{FM} = v'^{2/3}_g \exp(2\ln^2\sigma),$$
$$\eta_C = \frac{4K_{n_1}}{3} v'^{1/3}_g\exp\left(\frac{1}{2}\ln^2\sigma\right). \tag{21}$$

Second aerosol moment:

$$\frac{\partial V_2}{\partial \theta} = - v_{zl}\frac{\partial V_2}{\partial \bar{z}} + I'k^{*2} + 2\varepsilon(S-1)V + 2\zeta V^2, \tag{22}$$

where

$$\frac{1}{\varepsilon} = \frac{1}{\varepsilon_{FM}} + \frac{1}{\varepsilon_C}, \qquad \frac{1}{\zeta} = \frac{1}{\zeta_{FM}} + \frac{1}{\zeta_C},$$

$$\varepsilon_{FM} = v'^{2/3}_g \exp(8\ln^2\sigma), \qquad \varepsilon_C = \frac{4K_{n_1}}{3} v'^{1/3}_g \exp\left(\frac{7}{2}\ln^2\sigma\right),$$

$$\zeta_{FM} = r'^{1/2}_g b_2 \exp(\tfrac{3}{2}\ln^2\sigma)[\exp(\tfrac{25}{8}\ln^2\sigma)$$
$$+ 2\exp(\tfrac{5}{8}\ln^2\sigma) + \exp(\tfrac{1}{8}\ln^2\sigma)],$$

$$\zeta_C = K[1 + \exp(\ln^2\sigma) + B_5(K_{n_1}/r'_g)$$
$$\times \exp(-\tfrac{1}{2}\ln^2\sigma)(1 + \exp(-2\ln^2\sigma))]. \tag{23}$$

Expressing t and z in Eq. (2) in dimensionless variables and defining a new vector of state variables $x = [N\; V\; V_2\; \bar{x}]$, the approximate PDE system of Eqs. (18), (20) and (22) and the system of Eq. (2) (which describes the spatio-temporal evolution of concentrations of species and temperature of the continuous phase) yield the following general first-order hyperbolic PDE system:

$$\frac{\partial x}{\partial \theta} = A\frac{\partial x}{\partial \bar{z}} + f(x) + g(x)b(\bar{z})u(\theta), \tag{24}$$

where the specific form of the matrix A and the nonlinear vector functions $f(x)$, $g(x)$ is omitted due to space limitations. The system of Eq. (24) will be used for controller design in the next section.

Remark 2. The computation of the fractional moments in Eqs. (11), (13) and (14) and Eqs. (15)–(17) can be performed by expressing the fractional moments in terms of M_0, M_1, M_2 using the relation

$$M_k = M_0 v^k_g \exp\left(\frac{9}{2} k^2\ln^2\sigma\right)$$
$$= M_0\left(\frac{M_1^2}{M_0^{3/2}M_2^{1/2}}\right)^k \exp\left(\frac{1}{2}k^2\ln\left(\frac{M_0 M_2}{M_1^2}\right)\right), \tag{25}$$

which was derived by substituting the lognormal size distribution of Eq. (8) into Eq. (9) and using the expressions for v_g and $\ln^2\sigma$ of Eq. (10).

Remark 3. The rate of change of S can be obtained from a monomer balance and is given by:

$$\frac{\partial S}{\partial \theta} = - v_{zl}\frac{\partial S}{\partial \bar{z}} + R'_r - I'k^* - \eta(S-1)N. \tag{26}$$

4. Nonlinear feedback controller design

In this section, we synthesize nonlinear distributed output feedback controllers for spatially inhomogeneous aerosol processes described by Eqs. (1) and (2) that achieve aerosol size distributions with desired characteristics. The controller design problem will be addressed on the basis of the hyperbolic PDE system of Eq. (24). Such systems are characterized by spatial differential operators whose eigenvalues cluster along vertical or nearly vertical asymptotes in the complex plane, which implies that an infinite number of eigenmodes is required to accurately describe their dynamic behavior. This prohibits the application of modal decomposition techniques to derive ODE models that approximately describe the dynamics of the PDE system and suggests addressing the control problem directly on the basis of the PDE system. The geometric control methodology developed in Christofides and Daoutidis (1996) will be employed for controller design (the reader may refer to Christofides and Daoutidis (1996) for details and proofs of the results).

We initially define a general controlled output, $y(\theta)$, as

$$y(\theta) = \int_0^1 c(\bar{z})h(x(\bar{z}, \theta))\,\mathrm{d}\bar{z} \equiv \mathscr{C}h(x), \tag{27}$$

where $h(x(\bar{z}, \theta))$ is a nonlinear function of the state of the process (this allows considering controlled outputs that depend in a nonlinear fashion of x, e.g. v_g), $c(\bar{z})$ is a smooth known function which depends on the desired control objective (for example, regulation of v_g in the outlet of an aerosol reactor requires $c(\bar{z}) = \delta(\bar{z} - 1)$), and $\mathscr{C}$ is an integral operator. The definition of controlled output allows introducing the concept of characteristic index between the output y and the manipulated input u which will be used in the synthesis of the controller. More specifically, referring to the system of Eq. (24), the characteristic index of y with respect to u is the smallest integer σ for which (Christofides and Daoutidis, 1996):

$$\mathscr{C}L_g\left(\sum_{j=1}^n \frac{\partial x_j}{\partial \bar{z}} L_{a_j} + L_f\right)^{\sigma-1} h(x)\, b(\bar{z}) \not\equiv 0, \tag{28}$$

where a_j denotes the jth column vector of the matrix $A(x)$, and L_{a_j}, L_f denote the standard Lie derivative notation ($L_f h(x) = (\partial h/\partial x)f(x)$). From the above definition, it follows that σ depends on the structure of the process (matrix A and functions $f(x), g(x), h(x)$), as well as on the actuator and performance specification functions, $b(\bar{z})$ and $c(\bar{z})$, respectively.

The state feedback control problem is formulated as the one of synthesizing distributed controllers of the general form

$$u = \mathscr{S}(x) + s(x)\, y_{sp}, \tag{29}$$

where $\mathscr{S}(x)$ is a smooth nonlinear operator, $s(x)$ is an invertible matrix of smooth functionals, and y_{sp} is the set-point, that stabilize the closed-loop system and force the following linear input/output response:

$$\gamma_\sigma \frac{\mathrm{d}^\sigma y}{\mathrm{d}\theta^\sigma} + \cdots + \gamma_1 \frac{\mathrm{d}y}{\mathrm{d}\theta} + y = y_{sp}, \tag{30}$$

where $\gamma_1, \gamma_2, \ldots, \gamma_\sigma$ are adjustable parameters which can be chosen to guarantee input/output stability in the closed-loop system.

This controller synthesis problem leads to the following nonlinear distributed state feedback controller (Christofides and Daoutidis, 1996):

$$u = \left[\mathscr{C}\gamma_\sigma L_g\left(\sum_{j=1}^n \frac{\partial x_j}{\partial \bar{z}} L_{a_j} + L_f\right)^{\sigma-1} h(x)b(\bar{z})\right]^{-1}$$
$$\times \left\{y_{sp} - \mathscr{C}h(x) - \sum_{v=1}^\sigma \mathscr{C}\gamma_v\left(\sum_{j=1}^n \frac{\partial x_j}{\partial \bar{z}} L_{a_j} + L_f\right)^v h(x)\right\}, \tag{31}$$

which enforces stability and the response of Eq. (30) in the closed-loop system of Eqs. (24)–(31), provided that the the system of Eq. (24) is minimum-phase (the reader may refer to Christofides and Daoutidis (1996) for a rigorous definition of the concept of minimum-phase for hyperbolic PDE systems).

The implementation of the distributed nonlinear controller of Eq. (31) requires knowledge of the state $x(\bar{z}, \theta)$ at all positions and times, which may not be available in some practical applications. To overcome this problem, we use the following state observer to estimate the state $x(\bar{z}, \theta)$ of the system of Eq. (24) in space and time:

$$\frac{\partial \omega}{\partial \theta} = A\frac{\partial \omega}{\partial \bar{z}} + f(\omega) + g(\omega)\, b(\bar{z})u + \mathscr{P}(y - \mathscr{C}h(\omega)), \tag{32}$$

where ω denotes the observer state vector and $\mathscr{P}$ is a linear operator, designed on the basis of the linearization of the system of Eq. (32) so that the system of Eq. (32) is exponentially stable.

The state observer of Eq. (32) can be coupled with the state feedback controller of Eq. (31) to derive a nonlinear distributed output feedback controller of the form

$$\frac{\partial \omega}{\partial \theta} = A\frac{\partial \omega}{\partial \bar{z}} + f(\omega) + g(\omega)b(\bar{z})$$
$$\times \left[\gamma_\sigma \mathscr{C}L_g\left(\sum_{j=1}^n \frac{\partial \omega_j}{\partial \bar{z}} L_{a_j} + L_f\right)^{\sigma-1} h(\omega)b(\bar{z})\right]^{-1}$$
$$\times \left\{y_{sp} - \mathscr{C}h(\omega) - \sum_{v=1}^\sigma \gamma_v \mathscr{C}\right.$$
$$\left. \times \left(\sum_{j=1}^n \frac{\partial \omega_j}{\partial \bar{z}} L_{a_j} + L_f\right)^v h(\omega)\right\} + \mathscr{P}(y - \mathscr{C}h(\omega)),$$

$$u = \left[\gamma_\sigma \mathscr{C} L_g \left(\sum_{j=1}^{n} \frac{\partial \omega_j}{\partial \bar{z}} L_{a_j} + L_f \right)^{\sigma-1} h(\omega) b(\bar{z}) \right]^{-1}$$

$$\times \left\{ y_{sp} - \mathscr{C}h(\omega) - \sum_{v=1}^{\sigma} \gamma_v \mathscr{C} \left(\sum_{j=1}^{n} \frac{\partial \omega_j}{\partial \bar{z}} L_{a_j} + L_f \right)^{v} h(\omega) \right\}$$

$$(33)$$

that enforces stability and the input/output response of Eq. (30) in the closed-loop system of Eqs. (24)–(33).

Remark 4. The calculation of the control action from the controllers of Eqs. (31)–(33) requires algebraic manipulations as well as differentiations and integrations in space, which is expected because of their distributed nature.

Remark 5. Note that in the case of imperfect initialization of the observer states (i.e., $\omega(\bar{z}, 0) \neq x(\bar{z}, 0)$), although a slight deterioration of the performance may occur, (i.e., the input/output response of Eq. (30) will not be exactly imposed in the closed-loop system), the distributed output feedback controller of Eq. (33) guarantees exponential stability and asymptotic output tracking in the closed-loop system.

Remark 6. The exponential stability of the closed-loop system guarantees that in the presence of small errors in process parameters, the states of the closed-loop system will be bounded. Furthermore, one can also couple the nonlinear controller of Eq. (31) (Eq. (33)) with a proportional integral controller (i.e., substitute $y_{sp} - \mathscr{C}h(x)$ ($y_{sp} - \mathscr{C}h(\omega)$) by $y_{sp} - y + (1/\tau)\xi_p$, where ξ_p is the integral controller state, which is governed by $\dot{\xi}_p = y_{sp} - y$, $\xi_p(0) = 0$ to ensure asymptotic offsetless output tracking in the closed-loop system, in the presence of constant unknown process parameters and unmeasured disturbance inputs.

5. Simulation study

In this section, we describe an application of the proposed nonlinear control method to an aerosol flow reactor used to produce NH_4Cl particles. (see Dahlin et al. (1981) for further discussion on such a reactor.) The following chemical reaction takes place $NH_3 + HCl \rightarrow NH_4Cl$ where NH_3, HCl are the reactant species and NH_4Cl is the monomer species. Under the assumption of lognormal aerosol size distribution, the mathematical model that describes the evolution of the first three moments of the distribution, together with the monomer (NH_4Cl) and reactant (NH_3, HCl) concentrations and reactor temperature takes the form

$$\frac{\partial N}{\partial \theta} = -v_{zl} \frac{\partial N}{\partial \bar{z}} + I' - \xi N^2,$$

$$\frac{\partial V}{\partial \theta} = -v_{zl} \frac{\partial V}{\partial \bar{z}} + I'k^* + \eta(S-1)N,$$

$$\frac{\partial V_2}{\partial \theta} = -v_{zl} \frac{\partial V_2}{\partial \bar{z}} + I'k^{*2} + 2\varepsilon(S-1) V + 2\zeta V^2,$$

$$\frac{\partial S}{\partial \theta} = -v_{zl} \frac{\partial S}{\partial \bar{z}} + C\bar{C}_1\bar{C}_2 - I'k^* - \eta(S-1)N, \qquad (34)$$

$$\frac{\partial \bar{C}_1}{\partial \theta} = -v_{zl} \frac{\partial \bar{C}_1}{\partial \bar{z}} - A_1\bar{C}_1\bar{C}_2,$$

$$\frac{\partial \bar{C}_2}{\partial \theta} = -v_{zl} \frac{\partial \bar{C}_2}{\partial \bar{z}} - A_2\bar{C}_1\bar{C}_2,$$

$$\frac{\partial \bar{T}}{\partial \theta} = -v_{zl} \frac{\partial \bar{T}}{\partial \bar{z}} + B\bar{C}_1\bar{C}_2\bar{T} + E\bar{T}(\bar{T}_w - \bar{T}),$$

where $\bar{C}_1$ and $\bar{C}_2$ are the dimensionless concentrations of NH_3 and HCl, respectively, $\bar{T}$, $\bar{T}_w$ are the dimensionless reactor and wall temperatures, respectively, and A_1, A_2, B, C, E are dimensionless quantities. The explicit expressions of A_1, A_2, B, C, E are given in Table 2 and the values of the process parameters used in the simulations are given in Table 3.

Fig. 2 displays the steady-state profile of the dimensionless total particle concentration, N, as a function of dimensionless reactor length. As expected, N increases very fast close to the inlet of the reactor (approximately, the first 3% of the reactor) due to a nucleation burst, and then, it slowly decreases in the remaining part of the reactor due to coagulation. Note that even though coagulation decreases the total number of particles, it leads to the formation of bigger particles, and thus, it increases the geometric average particle volume, v_g.

We formulate the control problem as the one of controlling the geometric average particle volume in the outlet of the reactor, $v_g(1, \theta)$, ($v_g(1, \theta)$ is directly related to the geometric average particle diameter, and hence, it is a key product characteristic of industrial aerosol processes) by manipulating the wall temperature, i.e.

$$y(\theta) = \mathscr{C}v_g = v_g(1, \theta), \qquad u(\theta) = \bar{T}_w(\theta) - \bar{T}_{ws}, \qquad (35)$$

where $\mathscr{C}(\cdot) = \int_0^1 \delta(\bar{z} - 1)(\cdot)\,d\bar{z}$ and $\bar{T}_{ws} = T_{ws}/T_0 = 1$. Since coagulation is the main mechanism that determines the size of the aerosol particles, we focus on controlling

Table 2
Dimensionless variables for the model of Eq. (34)

$A_1 = \tau k P_0 y_{20}/RT_0$
$A_2 = \tau k P_0 y_{10}/RT_0$
$B = P_0 k \tau \Delta H_R y_{10} y_{20}/RT_0^2 C_p$
$C = N_{av} k \tau y_{10} y_{20} (P_0/RT_0)^2/n_{s0}$
$E = 4URT_0\tau/DC_pP_0$
$\bar{C}_i = y_i/y_{i0}\bar{T}$
$\bar{T} = T/T_0$
$\bar{T}_w = T_w/T_0$

Table 3
Process model parameters for the simulation study

$L = 20$ m	Reactor length
$D = 0.05$ m	Reactor diameter
$Re = 2000$	Reynolds number
$P_0 = 1$ atm	Process pressure
$T_0 = 298$ K	Inlet temperature
$y_{10} = y_{20} = 40$ ppm	Inlet mole fraction of reactants
$U = 10.4 \, \text{J m}^{-2} \, \text{s}^{-1} \, \text{K}^{-1}$	Overall coefficient of heat transfer
$\Delta H_R = 175.7 \, \text{K J mol}^{-1}$	Heat of reaction
$C_p = 29.1 \, \text{J mol}^{-1} \, \text{K}^{-1}$	Heat capacity of process fluid
$MW_g = 14.0 \times 10^{-3} \, \text{kg mol}^{-1}$	Mol. wt. of process fluid
$k = 11.4 \, \text{m}^3 \, \text{mol}^{-1} \, \text{s}^{-1}$	Reaction constant
$\mu = 3.5 \times 10^{-6} \, \text{kg m}^{-1} \, \text{s}^{-1}$	Viscosity of process fluid
$MW = 53.5 \times 10^{-3} \, \text{kg mol}^{-1}$	Mol. wt. of condensable species
$\log P_s \, (\text{mm Hg}) = -4644/T + 0.906 \log T - 0.00162T + 9.004$	Vapor pressure–temperature relation
$\gamma = 0.08 \, \text{N m}^{-1}$	Surface tension
$v_1 = 5.33 \times 10^{-29} \, \text{m}^3$	Monomer volume
$R = 8.314 \, \text{J mol}^{-1} \, \text{K}^{-1}$	Universal gas constant
$N_{av} = 6.023 \times 10^{23} \, \# \, \text{mol}^{-1}$	Avogadro's constant
$k_B = 1.38 \times 10^{-23} \, \text{J K}^{-1}$	Boltzmann's constant
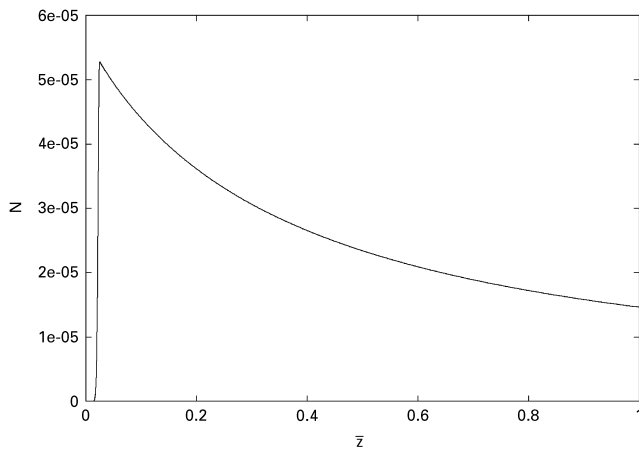
Fig. 2. Steady-state profile of dimensionless particle concentration.

Fig. 3. Closed-loop profiles of v_g in the outlet of the reactor under nonlinear and proportional integral controllers.

the part of the reactor where coagulation occurs. Therefore, the wall temperature is assumed to be equal to its steady-state value in the first 3.5% of the reactor (where nucleation mainly occurs), and it is adjusted by the controller in the remaining part of the reactor (where coagulation takes place).

The model of Eq. (34) was used as the basis for the synthesis of a nonlinear controller utilizing the proposed control method. For this model, σ was found to be equal to 2 and the necessary controller was synthesized using the formula of Eq. (31) and is of the form:

$$u = \left[\mathscr{C} \gamma_\sigma L_g \left(\sum_{j=1}^{n} \frac{\partial x_j}{\partial \bar{z}} L_{a_j} + L_f \right) h(x) b(\bar{z}) \right]^{-1}$$

$$\times \left\{ y_{sp} - \mathscr{C} h(x) - \sum_{v=1}^{2} \mathscr{C} \gamma_v \left(\sum_{j=1}^{n} \frac{\partial x_j}{\partial \bar{z}} L_{a_j} + L_f \right)^v h(x) \right\},$$
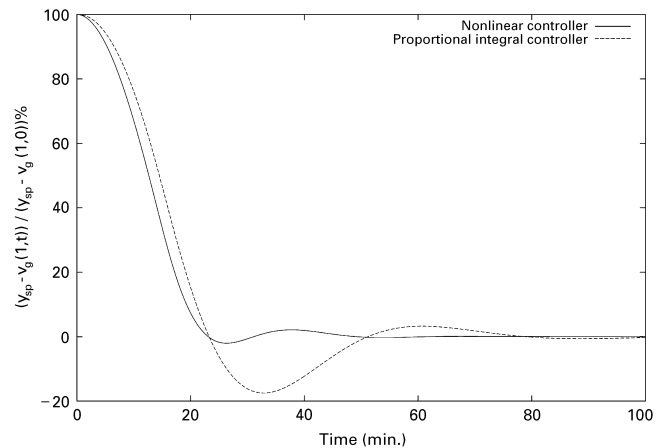
$$(36)$$

where $\gamma_1 = 580$ and $\gamma_2 = 1.6 \times 10^5$, to enforce a slightly underdamped response. Note that since the control objective is to regulate v_g in the outlet of the reactor (and therefore, $\mathscr{C}(\cdot) = \int_0^1 \delta(\bar{z} - 1) \ (\cdot) \, d\bar{z}$), the practical implementation of the controller of Eq. (36) requires only measurements of the process variables at $\bar{z} = 1$ (outlet of the reactor), which can be readily obtained in practice.

Two simulation runs were performed to evaluate the set-point tracking capabilities of the nonlinear controller and compare its performance with the one of a proportional integral controller. In all the simulation runs, the aerosol reactor was initially assumed to be at steady-state and a 5% increase in the set-point value of $v_g(1, 0)$ was imposed at $t = 0$ s (i.e. $y_{sp} = 1.05 v_g(1, 0)$). Fig. 3 and 4 show the controlled output and manipulated input profiles, respectively, of the nonlinear controller, and compare them with the corresponding profiles of
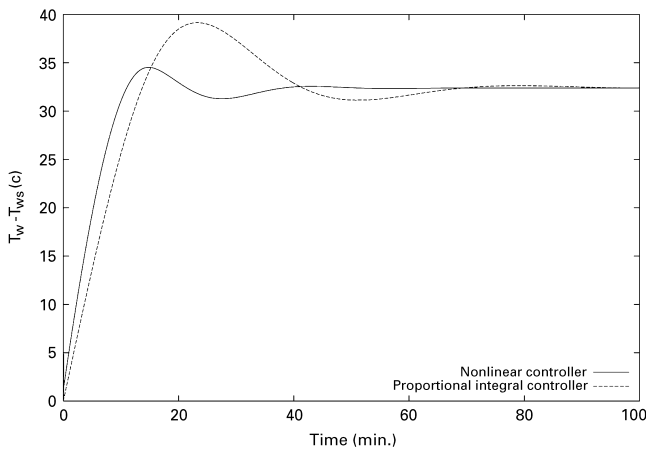
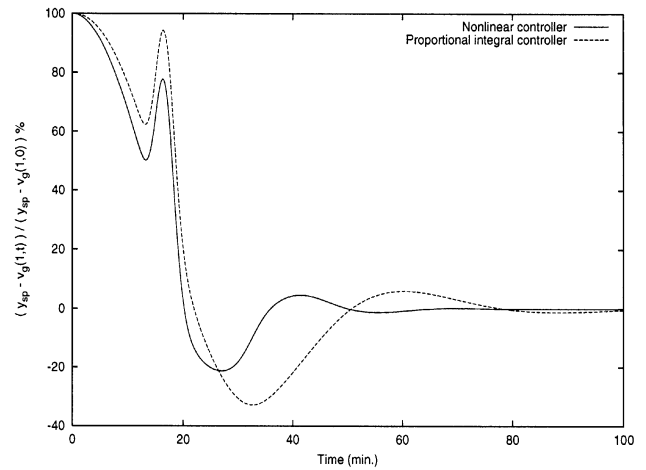Fig. 4. Manipulated input profiles for nonlinear and proportional integral controllers.

Fig. 6. Closed-loop profiles of v_g in the outlet of the reactor under nonlinear and proportional integral controllers in the presence of disturbance in the feed temperature.
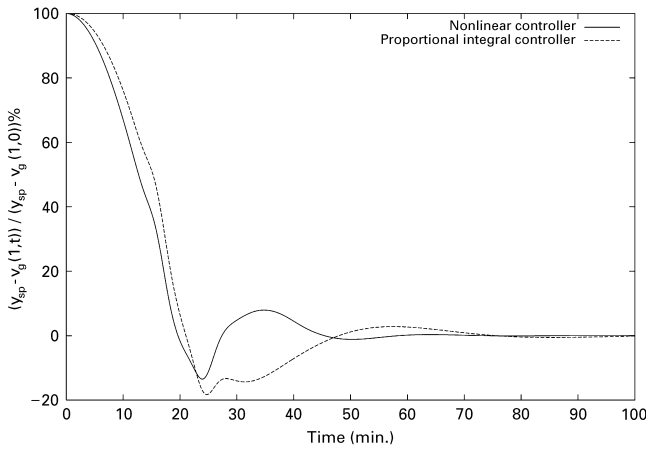

Fig. 5. Closed-loop profiles of v_g in the outlet of the reactor under nonlinear and proportional integral controllers in the presence of disturbance in NH$_3$ feed concentration.

Remark 7. Regarding the practical implementation of the nonlinear distributed controller of Eq. (36), we note that the wall temperature, $\bar{T}_w$ is not manipulated directly, but indirectly through manipulation of the jacket inlet flow rate. To this end, a controller should be designed based on an ODE model that describes the jacket dynamics, that operates in an internal loop to manipulate the jacket inlet flow rate to ensure that the jacket temperature obtains the values computed by the distributed robust controller (Christofides and Daoutidis, 1996). Of course, when such a controller is used, a slight deterioration of the closed-loop response obtained under the assumption that $\bar{T}_w$ can be manipulated directly, will occur.

6. Conclusions

In this work, we developed a nonlinear feedback control methodology for aerosol processes with simultaneous nucleation, condensation, coagulation and convective transport. Under the assumption that the aerosol size distribution is described by a lognormal function, the method of moments was initially employed to exactly reduce the population balance model into a set of three first-order hyperbolic PDEs which accurately describes the spatio-temporal evolution of the three leading moments of the aerosol size distribution. This system, together with the hyperbolic PDE system that describes the spatio-temporal evolution of the concentrations of species and temperature of the continuous phase, were used as the basis for the synthesis of nonlinear distributed output feedback controllers that use process measurements to attain an aerosol size distribution with desired characteristics. The control method was successfully

a proportional integral controller of the form

$$\dot{\xi}_p = y_{sp} - y, \qquad \xi_p(0) = 0,$$

$$u = K(y_{sp} - y) + \frac{1}{\tau}\xi_p \tag{37}$$

with $K = 2 \times 10^{15}$ and $\tau = 5 \times 10^{-8}$ (K and τ were computed after extensive trial and errors.). The proposed nonlinear controller regulates $v_g(1, \theta)$ successfully, to its new set-point value and outperforms the proportional integral controller. Figs. 5 and 6 show the controlled outputs in the wake of a 5% increase in the inlet concentration of NH$_3$ (from $t = 0$ to 500 s) and inlet temperature (from $t = 0$ to 100 s), respectively. Again, the nonlinear controller outperforms the proportional integral controller.

tested on a typical aerosol process and was shown to outperform a proportional integral control scheme and deal effectively with disturbances in the feed to the process.

Acknowledgements

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