

# Simulation and control of nanoparticle size distribution in a high temperature reactor

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This work focuses on the modeling, simulation and control of particle size distribution (PSD) during nanoparticle growth with the simultaneous chemical reaction, nucleation, condensation, coagulation and convective transport in a high temperature reactor. Firstly, a model known as population balance model was derived. This model describes the formation of particles via nucleation and growth. Mass and energy balances in the reactor were presented in order to study the effect of particle size distribution for each reaction mechanisms on the reactor dynamics, as well as the evolution of the concentrations of species and temperature of the continuous phase. The models were simulated to see whether the reduced population balance can be used to control the particle size distribution in the high temperature reactor. The simulation results from the above model demonstrated that the reduced population balance can be effectively used to control the PSD. The proposed method “which is the application of reduced population balance model” shows that there is some dependence of the average particle diameter on the wall temperature and the model can thus be used as a basis to synthesize a feedback controller where the manipulated variable is the wall temperature of the reactor and the control variable is the average particle diameter at the outlet of the reactor. The influence of disturbances on the average particle diameter was investigated and controlled to its new desired set point which is 1400nm using the proportional-integral-derivative controllers (PID controllers). The proposed model was used to control nanoparticle size distribution at the outlet of the reactor.

**Keywords:** Controller, nanoparticles, particle size, average particle diameter and wall temperature.

## INTRODUCTION

Nanoparticles are particles defined as a small object that behaves as whole in terms of its transport and properties. It is further classified according to size: in terms of diameter. Fine particles cover a range between 100 and 2500 nanometers, while ultrafine particles, on the other hand, are sized between 1 and 100 nanometers. Nanoparticles are of great scientific interest as they are effectively, a bridge between bulk materials and atomic or molecular structures. A bulk material should have constant physical properties regardless of its size, but at the nano-scale this is not often the case. Size-dependant properties such as quantum confinement in semiconductor particles, surface Plasmon resonance in some metal particles and superparamagnetism in magnetic materials, are observed. The properties of materials change as their size approaches the nanoscale and as the percentage of atoms at the surface of a material becomes significant. Nanoparticles always have novel properties that can be used for the development of the new improved processing, making the control synthesis of nanoparticles vital to fields like chemistry, materials science and engineering, and the environmental sciences. Again, nanosized (i.e., smaller than tens of nanometres) particles often behave differently, physically and chemically, from their larger counterparts. Thus, the synthesis of model materials for the fundamental mechanisms of nanoparticle transformations, nucleation and growth as well as the physical and chemical properties of nanoparticle are critical to understanding their behaviour in naturally engineered systems. Aerosol product such as  $\text{TiO}_2$  and  $\text{B}_4\text{C}$ , find widespread use as pigments, reinforcing agents, ceramic powders, optical fibres, carbon blacks and semiconductor

materials. Numerous experimental studies have suggested that aerosol growth occurs in stages, beginning with the gas phase chemical reaction of the reactants to produce monomers or molecules of the condensable species. The second stage is when monomers form unstable clusters, which further grow by monomer condensation. Beyond a critical cluster size, nucleation of stable aerosol particles occurs. These particles grow further by coagulations (condensation and surface reaction of some other growth mechanisms). “Particulate processes (also known as dispersed-phase processes) are characterized by material domains that are comprised of a continuous phase and are essential in making many high value industrial products. Examples include a nearly mono-disperse powder size distribution around 0.2–0.3  $\mu\text{m}$  which is required for the titania pigments so as to obtain the maximum opacity or hiding power<sup>1</sup>; the crystallization of proteins for pharmaceutical applications; the emulsion polymerization reactors for the production of latex; and thermal spray processing of nanostructured coatings”, Kalani and Christofides<sup>2</sup>. In aerosol processes, the growth or coagulation rate, which is affected additively by Brownian and turbulent shear forces strongly influences on particle size, size distribution and morphology; therefore, it is of fundamental interest in the synthesis of nanoparticles by aerosol processes. According to Kalani and Christofides<sup>2</sup>, it is now well-understood that the physico-chemical and mechanical properties of materials made with particulates depend heavily on the characteristics of the corresponding particle size distribution (PSD; for example, a nearly monodisperse PSD around 0.2–0.3  $\mu\text{m}$  is required for titania pigments to obtain the maximum hiding powder per unit mass. Numerical simulation provides a means of understanding and ultimately controlling the vari-

ous dynamics, transport, and mass and energy coupling processes in aerosol systems.

A high temperature reactor for producing spherical nanoparticles from vapor phase is a multi-composition system; varying appreciably in the flow and temperature fields. Nanoparticle synthesized in high temperature reactor often carries certain charges, which can be further utilized in the electrostatic manipulation, measurement and control of such particles. The nanoparticles produced at high temperature often undergo rapid coalescence, with complex rate laws, Michael and Markus<sup>3</sup>. "High-temperature methods based on flames or thermal plasma are often used for ultrafine (nanoscaled) powder synthesis. The main processes in these methods are the raw substance vapourization and the vapour-phase condensation that follows it. The condition under which the condensation takes place has an important impact on particle growth process", Balabanova<sup>4</sup>. There is a need for detailed understanding of nanoparticle formation especially the material synthesis and combustion in high temperature processes.

The dynamic models of aerosol processes are typically obtained from the application of population, material and energy balances and consist of partial integro-differential equation systems (where the independent variables are time, space and one or more internal particle coordinates, such as particle volume and shape). Nonlinearity usually arises from complex reaction, nucleation, 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. Numerical models can be viewed as mathematical frameworks that permit the interaction of complex physical processes to be simulated. The first step in developing a numerical aerosol model is to assemble expressions for the relevant physical processes, such as chemical reactions, nucleation, condensation, coagulation, etc. The next step is to approximate the particle size distribution with a mathematical size distribution function. The nonlinear and distributed nature of population balances have motivated extensive research on the development of efficient numerical methods for the accurate computation of their solution", Christofides et al<sup>5</sup>. Early research efforts had mainly focused on the understanding of the fundamental control theory properties. According to Christofides<sup>6</sup>, the recent efforts on model-based control of particulate processes have also been motivated by significant advances in modelling of particulate processes. From recent research in the dynamics of infinite-dimensional systems and nonlinear control theory, the control of particulate processes by applying the population balances has gradually developed into a very active research area within the field of process control in order to achieve tight particle size distribution. "For the past decade, research on model-based control has been limited despite the strong literature on population balance modelling, numerical solution, and dynamic analysis of particulate processes", Christofides et al<sup>5</sup>. In addition, there is a need to control these particulate processes in order to achieve tight control of particle size and its distributions.

Below are examples of solution methods.

## Sectional methods

Sectional methods (SM) are widely used to solve population equations. In these methods, the size spectrum is divided into a set of size classes. In so doing, distinction between zero-order and higher-order methods is made. Higher-order methods use low order polynomials to represent the particles within each section and can be regarded as a simple form of finite elements methods and will be discussed in the next section. They can suffer from stability problems and artificial dispersion; whereas zero-order methods are more robust. When using sectional methods, computational domain is divided into rather small intervals in which the solution is approximated by step functions. For each interval, obtained an ordinary differential equation which is coupled to neighbours depending on the discretization scheme used. Taluka and Swihart<sup>7</sup> used the sectional method to approximate the continuous size distribution by a finite number of sections or bins within which numerically conserved aerosol property is held constant. The particle number in each bin changes due to the contribution from coagulation, nucleation and growth. The method was used to treat coagulation based on conserving the mean volume of colliding particles. Xiong and Pratsinis quoted by Jeong and Choi<sup>8</sup> proposed a two-dimensional sectional model to solve the two-dimensional aerosol dynamics equation. They used a two dimensional particle size distribution function, in which particle volume and surface area are independent variables. The two dimension particle size domains, i.e., particle volume and surface area of particles are divided into a finite number of sections in which the two-dimensional aerosol dynamic equation can be reduced to a two-dimensional section population balance equation. Tsantilis et al<sup>9</sup> predicted the sectional model in terms of average diameters and geometric standard deviation, while Spicer et al<sup>10</sup> used a moving sectional population balance model. The change in the number of particles in the size section  $i$ ,  $N_i$ , by nucleation and coagulation was simulated. The predicted moving sectional model was evaluated for the evolution of an initially monodisperse size distribution by condensation only at constant growth rate.

## The least square method

The least squares method (LSM) is a well-established technique for solving a wide range of mathematical problems. The basic idea in the LSM is to minimize the integral of the square of the residual over the computational domain. In the case where the exact solutions are sufficiently smooth, the convergence rate is exponential. Daora and Jakobosen<sup>11</sup> applied the least square method to solve the population balance equation. Langston<sup>12</sup> compared the least squares method and the Bayes' theorem for deconvolution of mixture compositions.

## Monte Carlo methods

An alternative to sectional methods for solving the population balance equation are Monte Carlo (MC) methods. It is a very simple concept as it flows naturally from the conception of what a sampling distribution is. "The rule behind the Monte Carlo simulation is that the behaviour of a statistical concept in random samples can be estimated by the empirical process by

actually outlining lots of random samples and noticing this behaviour. The purpose for doing this is to create a pseudo-population, which looks like the real life situation in all relevant aspects". They are easy to implement, can account for fluctuations, and can easily incorporate several internal coordinates. The range of different physical phenomena which can be explored using Monte Carlo methods is exceedingly broad. Models which are obtained either naturally or through approximation can be discretized and considered, Landau and Binder<sup>13</sup>. In the case of nanoparticle modelling, the number of particles is so large that the fluctuations in particle numbers can be neglected. Mazo-Zuluaga et al<sup>14</sup> used Monte Carlo-Metropolis dynamic on the basis of a three dimensional classical Heisenberg model with the nearest magnetic neighbour interaction. They employed a single spin movement metropolis Monte Carlo algorithm for computing equilibrium thermodynamic properties. Monte Carlo steps per spin were considered in order to compute equilibrium averages. Iglesias and Labarta<sup>15</sup> also employed Monte Carlo simulation to simulate the magnetic properties of a model for a single nanoparticle consisting of ferromagnetic core that is surrounded by an antiferromagnetic shell. The model was performed for a nanoparticle consisting of Heisenberg spin on a simple cubic lattice. Efendiev and Zachariah<sup>16</sup> developed the hybrid Monte Carlo method for the simulation of simultaneous coagulation and phase segregation of an immiscible two-component binary aerosol. They described both the internal state of aerosol droplets and their growth. For the droplets, they employed a finite-number Monte Carlo and top up their system when the number of particles is halved. For the internal state of the droplets, they employed the finite-volume Monte Carlo approach and following each droplet, coagulation process was used to calculate the successful enclosure interaction in each droplet. The kinetic Monte Carlo (KMC) method is used to estimate and control methodologies for surface properties (e.g. surface roughness). Shi et al<sup>17</sup> developed Kinetic Monte-Carlo simulators of a surface based on small lattice size models in order to capture the dominant roughness evolution and utilize the available surface roughness measurements to improve on the predictions of the kinetic Monte-Carlo simulators in order to obtain accurate surface roughness estimates.

### Finite element methods

Alternative to sectional methods are the more sophisticated finite element methods. The finite element method is a numerical technique which gives approximate solutions to differential equations to problems arising in physics and engineering, Pepper and Heinrich<sup>18</sup>. In the finite element approach, the solution of the population balance is expanded in a series of polynomials. For the coefficients of this expansion, a set of equations has to be solved and this is obtained by inserting the expansion into the population balance equation. Various methods can be derived by different nodes, functions, and time stepping schemes. The mathematical discipline of functional analysis provides the theoretical framework with which errors can be estimated. This is of course a very attractive feature of finite element methods.

James and Christofides<sup>19</sup> proposed a finite-dimensional

approximation and control of nonlinear parabolic partial differential equation (PDE) systems by combining Galerkin's method with the concept of approximate initial manifolds, known as the non-linear Galerkin's method. Armaou and Christofides<sup>20</sup> employed some methodologies for the derivation of finite dimensional approximations and synthesis of low-dimensional nonlinear output feedback controllers for systems of nonlinear parabolic partial differential equation in spatial dimension with state description. Galerkin's method was used to derive low dimensional dynamic systems of nonlinear ordinary differential equation (ODE) that accurately reproduces the dynamics and solution of the nonlinear parabolic PDE system. Roussos, Alexopoulos and Kiparissides<sup>21</sup> used Galerkin's on finite elements method (GFEM) and moving grid technique to the numerically solved dynamic population balance in both the continuous and its discrete equivalent. The continuous form of the general balance equation was solved using GFEM. Accordingly, the volume domain is first discretized into "*ne*" elements, each element containing "*np*" nodal points. Subsequently, the number density function,  $n(v, t)$ , is approximated over each element in terms of its respective values at the nodal points using standard Lagrange interpolation polynomials.

### Methods of moments

The method of moments (MOM) is computationally the most efficient approach to obtain a numerical approximation to the moments of population balance. For this reason, this method is often used when simulating problems, where transport of particles in a flow with complex geometry is essential. In the area of nanoparticle modelling, two techniques have been used so far. One of the techniques is the quadrature method of moments (QMOM) which is a more recent technique. Dorao and Jakobsen<sup>22</sup> derived (QMOM) in two ways, i.e., the standard quadrature method of moments which is a numerical closure for method of moments (MOM) and the method of moments in the method of weighted residuals (MOM-MWR).

The second technique is the method of moment with internal closure (MOMIC). Diemer and Ehrman<sup>23</sup> developed a design for the comparison of reconstructed distributions from the moment with a direct calculation via the sectional method. The design was used to probe the sensitivity of distribution reconstruction and problem solution to the model size for the MOMIC and QMOM approaches. Barrett and Webb<sup>24</sup> compared some approximation methods for solving the aerosol dynamic equation. The methods compared are the quadrature method of moments, the finite element method (FEM), Laguerre quadrature and Associated Laguerre quadrature. In the first instance MOMIC has been developed to describe the formation and oxidation of soot particles. In its early form, the method is based on univariate description of spherical soot particles in the free molecular regime. An alternative approach to obtain the moments of the PSD is the quadrature method of moments (QMOM). In this method, the moments are calculated, assuming the PSD can be represented as weighted multi-dimensional Dirac delta function. The weights and the nodes are then chosen to satisfy the transport equations for the

moments of the PSD. The advantage of this approach is that due to the choice of delta functions, there exists no closure problem. Gerber and Mousavi<sup>25</sup> applied the quadrature method of moments to the polydispersed droplet spectrum in transonic steam flows with primary and secondary nucleation. Wright et al<sup>26</sup> used the bivariate extension of the quadrature method of moments (QMOM) to efficiently model the dynamic of a population of inorganic nanoparticles undergoing simultaneous coagulation and particle sintering.

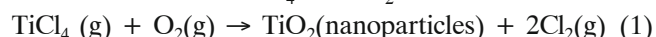
According to Williams and Loyalka quoted by Kalani and Christofides<sup>2</sup>, moment models provide adequate simplification by modelling the key average bulk properties of the evolving product, which may then be used to generate the approximate PSD under the assumption of a self-preserving form. The dominant dynamic behaviour of many aerosol processes can be accurately captured by a model that describes the evolution of the three leading moments of aerosol size distribution.

The objectives of this study are:

- To investigate the effect of wall temperature on the average particle diameter.
- To investigate the influences of disturbances on the average particle diameter.
- To control the average particle diameter to its new desired set point value.

## REACTION MODEL

Titanium dioxide (TiO<sub>2</sub>) particles are important in our daily lives. Titanium dioxide (TiO<sub>2</sub>) nano-particles are traditionally used as pigments, but have found use in diverse areas like photocatalysis and in reducing nitrogen oxide emissions. “The main route of titania nanoparticle production is the so-called chloride process. Raw titania is converted into titanium tetrachloride and subsequently this chloride is oxidised in a pure oxygen plasma at 1500K to 2000K, which leads to titania particles” Oliver and Markus<sup>27</sup>. The formation of TiO<sub>2</sub> takes place by the overall reaction of TiCl<sub>4</sub> with O<sub>2</sub>:



This research is focused on the multicomponent/multifunctional nanomaterial. This will increase the complexity of aerosol processes and the demands for their control. Process simulations via energy balance, mass balance and population balance modelling will provide a perspective for parameter studies, process and product optimisation, besides gaining a deeper understanding of the process consideration. However, the quality and amount of information of any result obtained from such simulation depends on the assumptions and approximations adopted.

## Population balance model

The population balance equation that consists of nonlinear partial integro-differential equation is given below.

$$\frac{\partial n}{\partial t} + \frac{\partial(G(v, z, \bar{x})n)}{\partial v} + c_z \frac{\partial n}{\partial z} - I(v^*)\delta(v - v^*) = \frac{1}{2} \int_0^v \beta(\tilde{v}, v - \tilde{v}, \bar{x})n(\tilde{v}, z, t)n(v - \tilde{v}, z, t)d\tilde{v} - n(v, z, t) \int_0^\infty \beta(v, \tilde{v})n(\tilde{v}, z, t)d\tilde{v} \quad (2)$$

The first term on the left hand side of Eq. (2) describes the change in the number concentration of particle volume interval  $v$ ,  $v+dv$  and in the spatial interval  $z$ ,  $z+dz$ ,  $n(v, z, t)$  denotes the particle size distribution

function,  $v$  is particle volume,  $t$  is time,  $z \in [0, L]$  is the spatial coordinate,  $L$  is the length of the process. The second term on the left hand side gives the loss or gain of particles by condensational growth, the third term on

the left hand side which is  $c_z \frac{\partial n}{\partial z}$  corresponds to the convective transport of aerosol particles at fluid velocity  $c_z$  and the fourth term on the left hand side accounts for the formation of new particles of critical volume  $v^*$  by nucleation rate  $I(v^*)\delta(v - v^*)$ , also accounts for gain and loss of particles by condensation.  $G(v, z, \bar{x})$ ,  $I(v^*)$  and  $\beta(\tilde{v}, v - \tilde{v}, \bar{x})$  are the nonlinear scalar functions and  $\delta$  is the standard Dirac function. The mass and energy balance model which predicts the spatio-temporal evolution of the concentrations of species and temperature of the gas phase given by Ashish and Panagiotis<sup>28</sup> has the following form:

$$\frac{d\bar{x}}{dt} = \bar{A} \frac{d\bar{x}}{dz} + \bar{f}(\bar{x}) + \bar{g}(\bar{x})b(z)u(t) + \tilde{A} \int_0^\infty a(\eta, v, x)dv \quad (3)$$

where  $\bar{x}(z, t)$  is an  $n$ -dimensional vector of state variables that depends 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 temperatures,  $T_{w1}$  and  $T_{w2}$ ) and  $b(z)$  is a known function which determines how control action  $u(t)$ , is distributed in space. The last term on the right hand side of Eq. (3) accounts for mass and heat transfer from the continuous phase to all the particles in the population. The gain and loss of particles by Brownian coagulation is described by the first and second term on the right hand side of Eq. (2) respectively.

$$\frac{1}{2} \int_0^v \beta(\tilde{v}, v - \tilde{v}, \bar{x})n(\tilde{v}, z, t)n(v - \tilde{v}, z, t)d\tilde{v} - n(v, z, t) \int_0^\infty \beta(v, \tilde{v})n(\tilde{v}, z, t)d\tilde{v} \quad (4)$$

## Mathematical model

The mathematical model that describes the evolution of the first three moments of distribution, together with the monomer and reactant concentration and temperature is given by Ashish and Christofides [2] and they take the following form.

$$\frac{dN}{d\theta} = -c_{zl} \frac{dN}{d\bar{z}} + I' - \xi N^2 \quad (5)$$

$$\frac{dV}{d\theta} = -c_{zl} \frac{dV}{d\bar{z}} + I'k^* + \eta(S-1)N \quad (6)$$

$$\frac{dV_2}{d\theta} = -c_{zl} \frac{dV_2}{d\bar{z}} + I'k^{*2} + 2\varepsilon(S-1)V + 2\zeta V^2 \quad (7)$$

$$\frac{dS}{d\theta} = -c_{zl} \frac{dS}{d\bar{z}} + C\bar{C}_1\bar{C}_2 - I'k^* - \eta(S-1)N \quad (8)$$

$$\frac{d\bar{C}_1}{d\theta} = -c_{zl} \frac{d\bar{C}_1}{d\theta} - A_1\bar{C}_1\bar{C}_2 \quad (9)$$

$$\frac{d\bar{C}_2}{d\theta} = -c_{zl} \frac{d\bar{C}_2}{d\theta} - A_2\bar{C}_1\bar{C}_2 \quad (10)$$

$$\frac{d\bar{T}}{d\theta} = -v_{zl} \frac{d\bar{T}}{d\bar{z}} + B\bar{C}_1\bar{C}_2\bar{T} + E\bar{T}(\bar{T}_w - \bar{T}) \quad (11)$$

Where  $\bar{C}_1$  and  $\bar{C}_2$  are the dimensionless concentrations of the reactants,  $\bar{T}_w$ ,  $\bar{T}$  are the dimensionless reactor and wall temperature, respectively,  $A_1$ ,  $A_2$ ,  $B$ ,  $C$ ,  $E$  are the dimensionless quantities. Table 1 gives the dimensionless variable while table 2 gives the process parameters used in the simulation. Dimensionless quantities for the model

of Eqs (5–11), according to Ashish and Panagiotis<sup>28</sup>. An advanced understanding of parameters that determine the formation of particle is crucial for the successful simulation of the process, hence choosing the meaningful range of input parameters should be done with great care. The initial parameters were chosen close to the data published in literature for the same process, Kalani & Christofides<sup>2</sup>.

$$A_1 = \tau k P_0 y_{20} / RT_0, \quad A_2 = \tau k P_0 y_{10} / RT_0$$

$$B = P_0 k \tau \Delta H_R y_{10} y_{20} / RT_0^2 C_P, \quad \bar{C}_i = y_i / y_{10} \bar{T}$$

$$C = N_{av} k \tau y_{10} y_{20} (P_0 / RT_0)^2 / n_{s0}, \quad E = 4UR T_0 \tau / DC_P P_0,$$

$$\bar{T} = T / T_0 \quad \text{and} \quad \bar{T}_w = T_w / T_0.$$

## CONTROL SYSTEM

The task of a control system is to ensure the stability of the process, to minimize the influence of disturbances and perturbations and to optimize the overall performance. The objective of this study is achieved by maintaining the average particle diameter close to its desired value using the set point which is fixed on time-dependent.

### Control Scheme

The control scheme in this study is used to reduce the influence of disturbances and to check the process response to set point. The control scheme used is the PID controller. The closed loop profile of the system under the influence of disturbances ( $\text{TiCl}_4$  and  $\text{O}_2$  flowrates) with PID controller is given in Fig. 1 below.

## ANALYSIS OF RESULTS AND DISCUSSION

In this section, the process models of Equations (5–11) were numerically solved by using the Simulink, a part of (Matlab) software environment. It is an interactive computing package for simulating and analysing differential equations, mathematical models and dynamic systems. The computation was done using a multistep solver known as ode 15s (stiff/NDF) solver, ODE 15s is a variable-order solver based on the numerical differentiation formulas (NDFs). Optionally it uses the backward differential formulas BDFs, Sergey<sup>29</sup>. Since the objective of this study is to control nanoparticle growth with desired particle distribution in a high temperature reactor, the effect of wall temperature on the average particle diameter was studied. It is a variable that could be used in industry to control the aerosol size distribution.

In this section, the effect of wall temperature on the average particle diameter is discussed, in order to synthesize nonlinear output feedback controllers for titanium aerosol reactor that attain size distributions with desired characteristics. The effect of wall temperature on the average particle diameter was investigated in the open-loop simulation. This is to enable the application of the model as a basis to synthesize feedback controllers which manipulated the input (wall temperatures) in order to achieve control of nanoparticle size and its distribution. From Fig.1, it was found that the particle size decreases with increasing wall temperature which clearly shows that the wall temperature is a variable that has some effects on the average particle diameter. The average particle diameter increases with increase in the dimensionless time. It can be seen from Fig. 2 below that

when particles are nucleated, a primary particle with a diameter of about 400nm is produced at constant time. After a certain number of particles have been produced, the frequency of bi-particle collision increases, resulting in a sharp increase in particle diameter. The reduction in dimensionless temperature is significant.

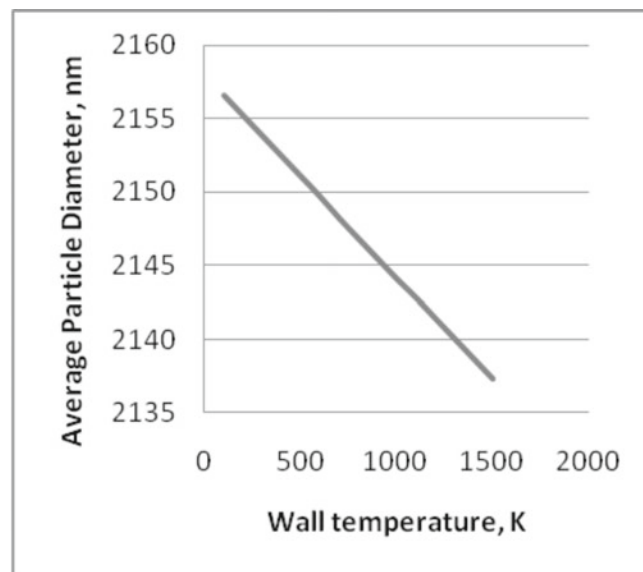


Figure 1. Average particle diameter as a function of wall temperature

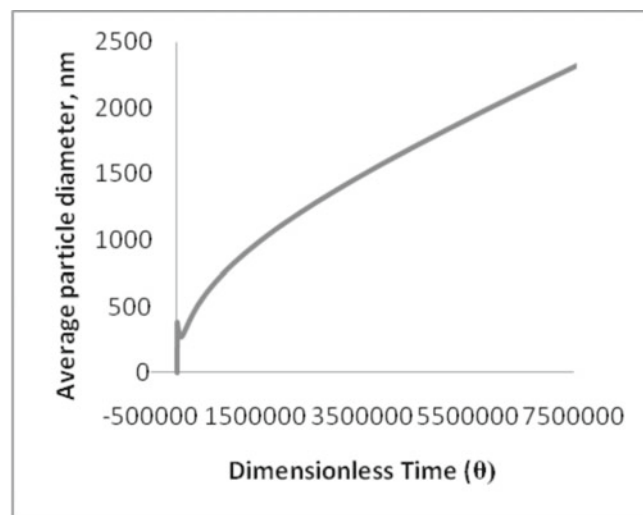


Figure 2. Average particle diameter as a function dimensionless time

Fig. 1 gives the average particle diameter as function of the wall temperature and Fig. 2 gives the average particle diameter as function of dimensionless time. The average particle diameter decreases as the wall temperature increases. This shows that the wall temperature has some effects on the average particle diameter.

It can be seen from Fig 3 that the saturation ratio initially accordingly, at about a dimensionless time of  $1e^6$ . Once the saturation ratio had increased to 1, some aerosol particles size increased due to coagulation causing an increase in the second aerosol moment as shown in Fig 4 and subsequently a reduction of saturation ratio in Fig. 3. During the descending stage, after sometime, the saturation ratio decreased continuously and dropped below 1. Several simulation runs were performed to investigate the effect of the disturbances of  $\text{TiCl}_4$ ,  $\text{O}_2$  flow rates on the average particle diameter with respect to

the model parameters. The disturbances actually affect the model because it made the system to be unstable. And because the process model is highly nonlinear, the influence of disturbance on the average particle diameter is non-stationary (random walk). Fig 5 shows the effect of disturbances of both  $\text{TiCl}_4$ ,  $\text{O}_2$  flow rates on the average particle diameter. It can be observed that disturbances have great effect on the outputs, thus the need to control particle size distribution.

Fig. 5 gives the average particle diameter under the influence of  $\text{TiCl}_4$  and  $\text{O}_2$  flowrates as disturbances while Fig. 6 gives the Comparison of the average particle

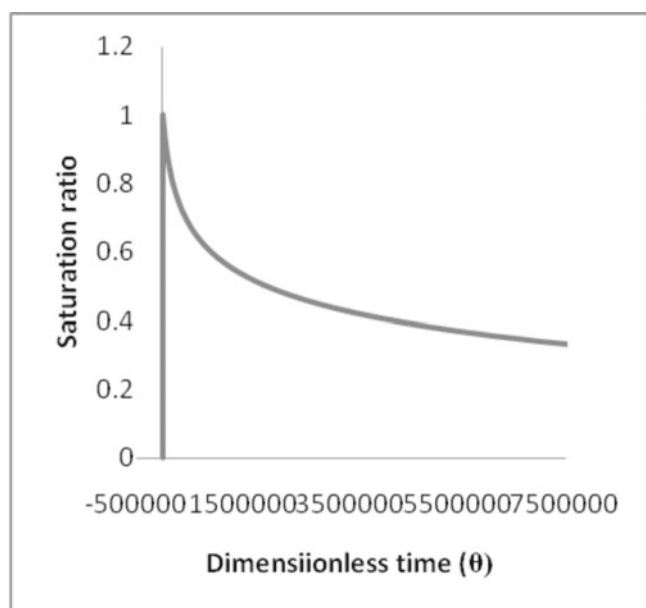


Figure 3. Saturation ratio as a function of dimensionless time

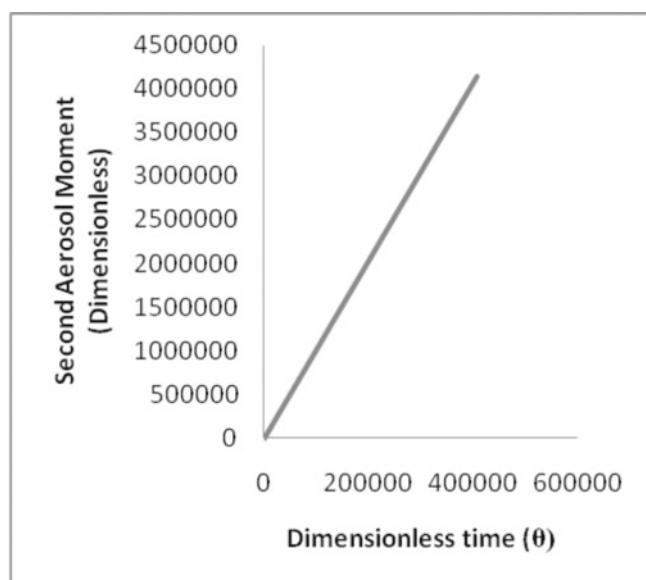


Figure 4. Second aerosol moment (dimensionless) as a function of dimensionless time

diameter with and without disturbance. From Fig. 6 above, the result of average particle diameter shows that the process is not distorted, while that of disturbance shows instability and this is attributed to the fact that the disturbance distorts the process.

Fig. 7 gives the closed loop profile of the system with PID controller while Fig. 8 gives the Simulink structure of the closed loop profile of the system with the PID controller. The PID controller compares the measured

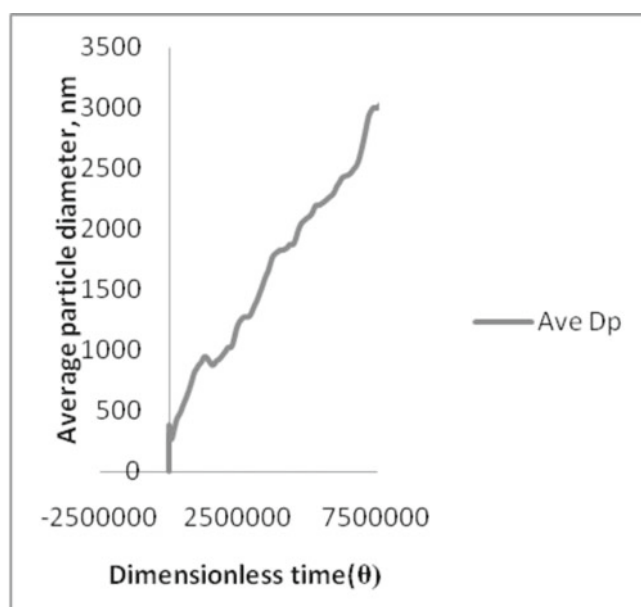


Figure 5. Average particle diameter under the influence of disturbances

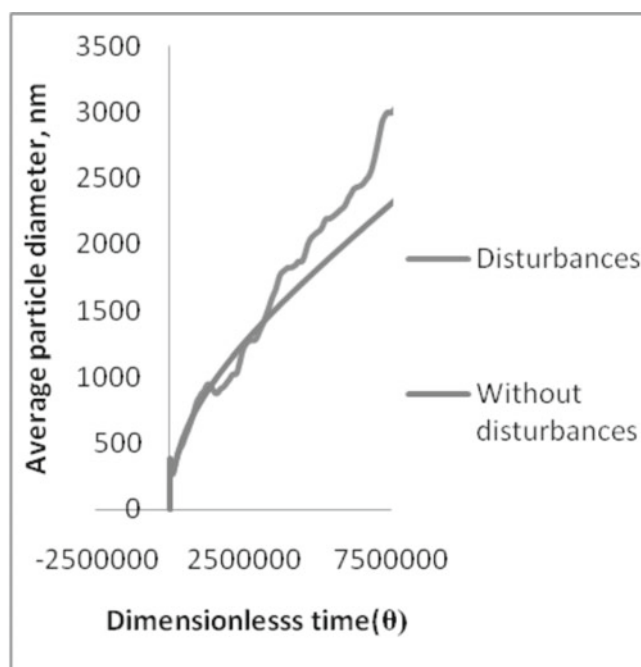


Figure 6. Comparison of average particle diameter with and without disturbances

process value  $Y$  with a reference set point. The error is then calculated to process a new process input  $u(t)$  which is the wall temperature  $T_w$ . This input will try and adjust the measured process value back to the desired set point.

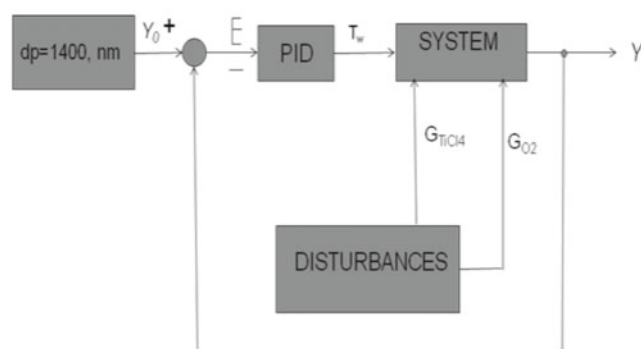


Figure 7. Closed loop profile of the system PID controller

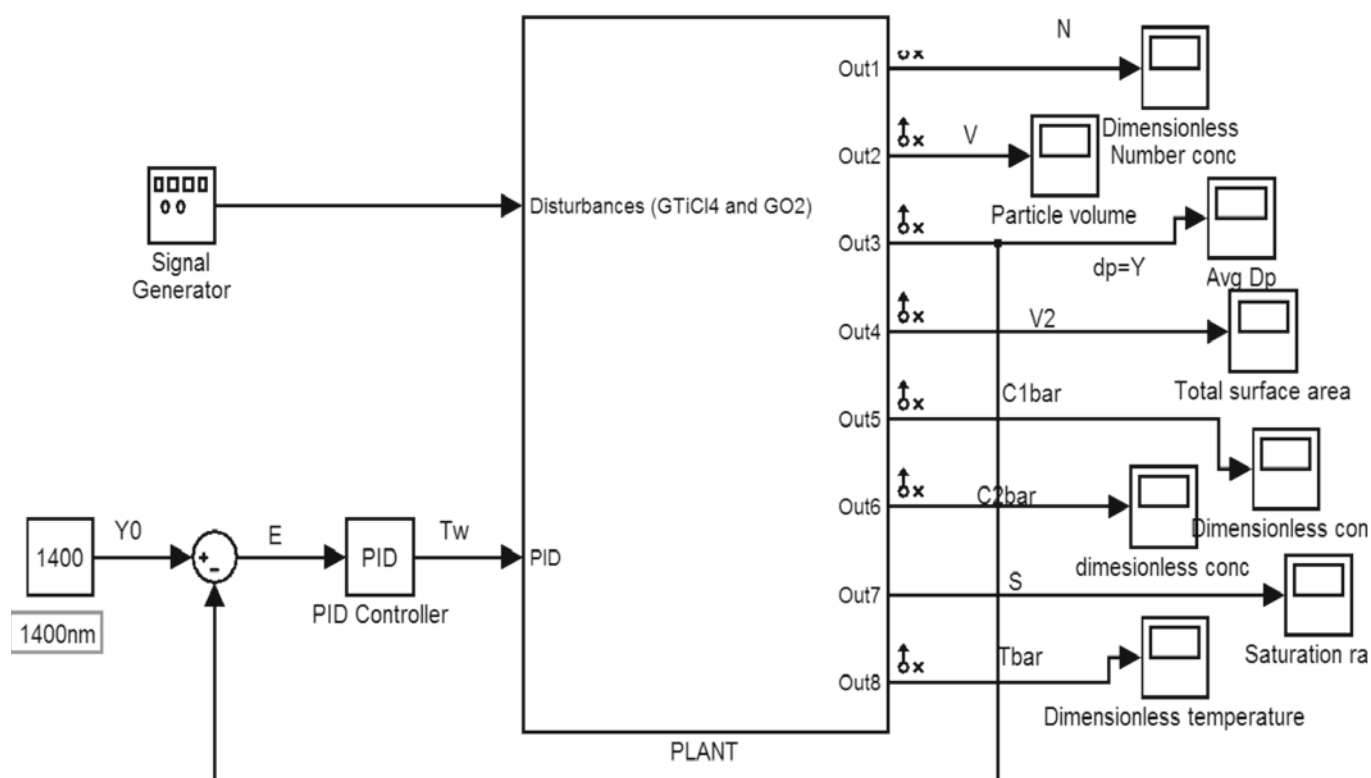


Figure 8. Simulink structure of the closed loop with profile of the system with PID controller

### Closed loop simulation

The process control configuration for the nanoparticle reactor was done through extensive “trial and error”. The control problem was formulated as one of tracking the average particle diameter of the aerosol system along a time-varying profile, by manipulating the wall temperature, i.e.  $y(t) = dp(t)$  and  $u(t) = T_w$ . The process model was investigated through the open loop simulation; hence it could be used to control particle size distribution. Therefore the model of equation (5–11) was used as the basis for the synthesis of feedback controller (proportional–integral controller) utilizing the proposed control method with  $k = 0.1$  and  $\tau = 4 \times 10^{-10}$ . ( $k$  and  $\tau$  were computed after extensive trial and errors). The process model parameters used for the simulation study are listed in Table 2, while Table 1 gives the dimensionless variables. The proposed controller regulates the particle diameter to attain the desire result to its new set point value by eliminating the effect of flowrate disturbances. This was done by manipulating the wall temperature (manipulated variable) in order to control the average

particle diameter (controlled variable). The error is then calculated in order to process a new process input  $u(t)$  which is the wall temperatures  $T_{w1}$  and  $T_{w2}$ . This input will try and adjust the measured process value back to the desired set point. Fig. 9 shows the controlled output of the average particle diameter (from  $t = 0$  to  $7.75e^6$ ). The wall temperature is not manipulated directly, but indirectly through the manipulation of wall temperature in the dimensionless temperature. To this end, the controller should be designed based on an ODE model that describes the dynamics that operates in an internal loop in order to manipulate the inlet flowrate and dimensionless temperature so as to ensure that the average particle diameter attains the values computed by the controller. The objective of the closed loop simulation is to show that the use of feedback control allows the production of an aerosol product with a desired average particle diameter ( $dp = 1400\text{nm}$ ). It is clear that the use of feedback control allows for the production of an aerosol product with a desired particle diameter ( $dp = 1400\text{nm}$ ).

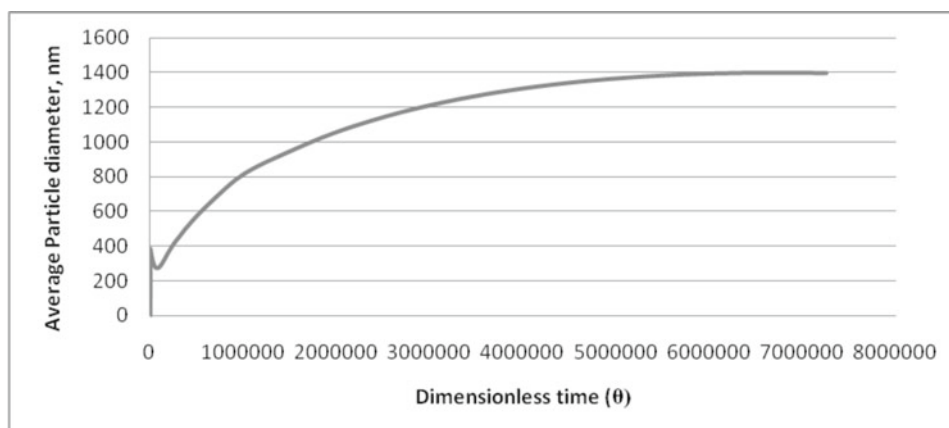


Figure 9. Closed – loop profiles of average particle diameter in the outlet of the reactor

**Table 1.** Dimensionless Variables by Ashish and Christofides<sup>2</sup>

$N = M_0 / n_s, V = M_1 / n_s v_1$	Aerosol concentration and volume
$V_2 = M_{2f} n_s v_1^2, K_{n_i} = \lambda / r_1$	Second aerosol moment and Knudsen number respectively
$\tau = (2 \pi m_{1/k} B T)^{1/2} n_s s_1, \bar{z} = z / L$	Characteristic time for particle growth and Dimensionless distance
$K = (2 k_B T / 3 \mu) n_s \tau, I' = I / (n_s / \tau)$	Coagulation coefficient and Nucleation rate
$v_g' = v_g / v_1, r_g' = r_g / r_1$	Dimensionless geometric volume and radius respectively
$c_{zi} = \tau c_z / L, \theta = t / \tau$	Dimensionless velocity and Dimensionless time respectively

**Table 2.** Process model parameters for the simulation study

$L = 1.5 \text{ m}, D = 0.1 \text{ m}$	Reactor length and Reactor Diameter respectively
$P^0 = 101000 \text{ pa}, T^0 = 2000 \text{ K}$	Process pressure and Inlet temperature respectively
$y^{10} = 0.4, y^{20} = 0.6$	Inlet mole fractions of $O_2$ and $TiCl_4$ respectively
$U = 160 \text{ J m}^{-2} \text{ s}^{-1} \text{ K}^{-1}, \Delta H_R = 88000 \text{ J mol}^{-1}$	Overall coefficient of heat transfer and Heat of reaction
$C_p = 1615.25 \text{ J mol}^{-1} \text{ K}^{-1}$	Heat capacity of the process fluid
$MW_g = 14.0 \times 10^{-3} \text{ kg mol}^{-1}, K = 11.4 \text{ m}^3 \text{ mol}^{-1} \text{ s}^{-1}$	Mol wt. of process fluid and Reaction rate constant
$\log P_s (\text{mmHg}) = -4644 / T + 0.906 \log T - 0.00162 T + 9.004$	PVT relation
$R = 8.314 \text{ J mol}^{-1} \text{ K}^{-1}, \sigma = 0.5$	Universal gas constant and Sigma respectively
$r_1 = 0.5e06 \mu\text{m}, r_g = 1.5e06 \mu\text{m}$	Monomer radius and Geometric radius respectively
$v_1 = 5.33 \times 10^{-29} \text{ m}^3, v_g = \pi d_p^3 / 6$	Monomer volume and Geometric volume respectively
$Re = 2000, T_w = 800 \text{ K}, 500 \text{ K}$	Reynolds number and Wall temperatures respectively
$\mu = 6.7 \times 10^{-5} \text{ kg m}^{-1} \text{ s}^{-1}$	Viscosity of process fluid
$s_0 = 1.1587 E - 19 \text{ m}, \gamma = 0.08 \text{ N m}^{-1}$	Monomer surface area and Surface tension respectively
$N_{av} = 6.023 \times 10^{23} \text{ # mol}^{-1}, k_B = 1.38 \times 10^{-23} \text{ J K}^{-1}$	Avogadro's and Boltzmann's constant

## CONCLUSIONS

In this work, the population balance model was developed. The model accounts for the simultaneous nucleation, condensation and coagulation for the control of size distribution. The effect of wall temperature on the average particle diameter was investigated in order to synthesize nonlinear output feedback controllers for titanium aerosol reactor that attains size distributions with desired characteristics. The average particle diameter can be decreased by increasing the reactor wall temperature. The performance of the open loop simulation was also validated with existing model and the results are identical. It was observed that the wall temperature has significant effect on the particle diameter. Based on the sensitivity of wall temperature upon the particle diameter,  $d_p$ , the process model sensitivity to the influence of disturbance was investigated. And it was observed that the process was affected by disturbances, thereby causing the process to be unstable. The process model was subsequently used as a basis to synthesize a feedback controller which manipulates the wall temperature of the reactor in order to control the aerosol size distribution in the outlet of the reactor with desired average particle diameter. The

controller reduced the effect of flowrate disturbances; it also reduced the effect of nonlinearity and distortion and it brings the process output to its desired set point value which is 1400nm.

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