# Modeling and simulation of nanoparticle production in an aerosol flame reactor

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## ABSTRACT

Aerosol flame synthesis is one of the commonly used methods for producing nanoparticles on a large scale. Particle size distribution (PSD) is one of the important variables that determines the end use of product nanoparticles. The PSD strongly depends 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, carrier gas and the burner geometry. A coupled flame dynamics-population balance model (PBM) for nanoparticle synthesis in an aerosol flame reactor is presented here. Various case studies for process design and scale up of a lab scale flame reactor and pilot scale furnace reactor for synthesis of titania, silica and carbon black will be presented. The model predictions were tested with published experimental data for flame temperature and particle size distributions. The model presented here will be useful for simulation of fine particle production in industrial furnace reactors and for studying design changes with respect to sizing of nozzles, location of heating elements and for identifying the correct location for introducing the coolant from the discharge end.

***Keywords***: aerosol flame reactor, nanoparticles, CFD, PBM, furnace reactor

## 1 INTRODUCTION

Flame aerosol synthesis is one of the commonly employed techniques for synthesis of various particulate commodities such as carbon black, ceramic powders (titania and fumed silica), speciality chemicals (zinc oxide and alumina) and several other high purity materials (advanced ceramics, semiconductors, super alloys and thin films etc.) [1-2]. Currently, this technology is being used to manufacture carbon black, titania, silica etc. on an industrial scale with annual production volumes of several million metric tons and particle production rates on the order of 100 metric tons per day [3-4] In this process, flame supplies the energy required to drive the chemical reactions (decomposition/pyrolysis) of the precursors. As a result, product monomers are formed which further grow to nanoparticles via intra-particle and inter-particle growth processes such as nucleation, surface growth, coagulation and coalescence [5]. Although this technique is well established on an industrial scale, very little is known about control and optimization of the large scale reactors due to extremely fast chemical reactions (~milliseconds) and complex particle growth processes under typical process conditions. To overcome the practical challenges, development of a model that can help in the optimal design and operation of these reactors is essential for producing particles with narrow size distribution and high specific surface area, besides desired chemical or phase composition.

The flame dynamics can be predicted using a computational fluid dynamics (CFD) model while population balance model (PBM) can be used to predict the particle size distribution with inputs from CFD model. In this work, we have coupled the CFD model and a one-dimensional sectional PBM for studying the effect of various process parameters on the product particle characteristics. This helps in control and optimization, design and scale up of the flame reactor for nanoparticle synthesis. The predictions of the coupled CFD-PBM model have been validated with experimental data for titania nanoparticle synthesis [6] in a pilot scale furnace reactor. The effects of key process parameters like furnace temperature and initial precursor concentration on the particle size distribution have also been studied. The details of the CFD [7] and PBM [8] models can be found elsewhere.

## 2 RESULTS AND DISCUSSION

A schematic of the furnace reactor used for the simulations is shown in Fig. 1(a). The carrier gas, precursor and oxygen are introduced through different pipes and are pre-mixed before entering the reactor. The reactor temperature is maintained by providing energy through an electrical furnace. Nitrogen is used as the coolant and it is introduced from the discharge end of the reactor. Details of the experimental procedures and operating conditions of the reactor can be found elsewhere [6].

The simulated temperature field inside the furnace reactor predicted by the commercial CFD package CFX 11.0 is shown in Fig. 1(b). It can be observed from Fig. 1(b) that the gas which is initially at room temperature, gets heated up in the region where the furnace heating elements are located, and then it is cooled upon mixing with the incoming nitrogen gas and exits the reactor. Towards the reactor exit, the gas temperature drastically reduces due to quenching with the incoming nitrogen gas. The temperature

of the gas stream is cooled down to below 600 K in the outlet product line. The experimental and predicted axial temperature profiles are shown in Fig.2. It can be noted that there is a good agreement between the experimental and predicted temperature profiles.

The particle size distribution is reported in terms of the dimensionless aggregate size distribution represented by $(\Delta N/N_T)/(\Delta \ln d_{sb})$ [8]. This quantity is defined as follows:

$$(\Delta N/N_T)\Big/(\Delta \ln d_{sb}) = \left(N_i \Big/ \sum_{i=1}^{M} N_i\right)\Big/\left(\ln d_{sb,i+1} - \ln d_{sb,i}\right) \quad (1)$$

Where $N_i$ is number concentration, $N_T$ is total number concentration, $d_{sb,i}$ is primary particle diamter in a section $i$.

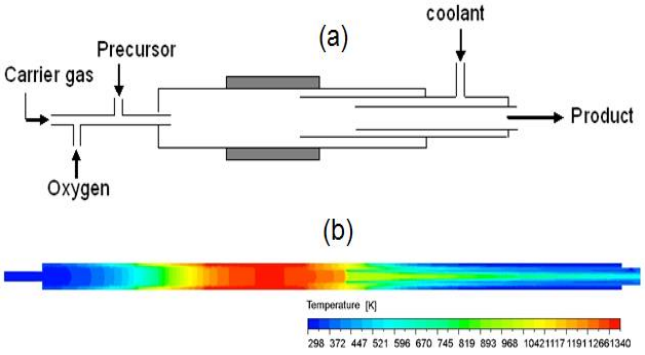


Figure 1: (a) Schematic of the furnace aerosol flame reactor; (b) Predicted temperature profile of the reactor.

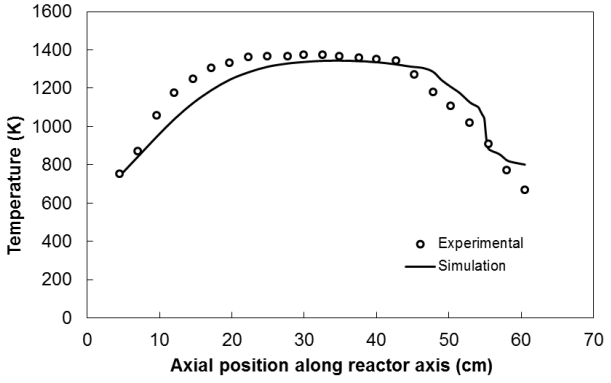


Figure 2: Comparision of predicted axial gas temperature profile of the furnace reactor with the experimentally measured profile [6] (Furnace set temperature: 1373 K).

Figs. 3(a) and 3(b) show the population balance model prediction of the aggregate size distribution at two different furnace set temperatures, 1600 K and 1723 K, respectively and their comparison with experimental data.

These results show the effect of change of furnace set temperature on the aggregate size distribution for an initial $TiCl_4$ concentration of $1.16\times10^{-5}$ mol/L. It can be observed that geometric number averaged diameter of the particles increases significantly as furnace set temperature increases. This can be attributed to the fact that reaction rate for formation of $TiO_2$ increases with increase in temperature. As a result, $TiO_2$ monomer concentration and thus collision frequency of the $TiO_2$ monomers significantly increases, which leads to the formation of bigger product particles at high temperatures. Fig. 4(a) and 4(b) show that the geometric mean size of the particles increases with an increase in the initial $TiCl_4$ concentration. At higher $TiCl_4$ concentrations, more titanium dioxide is produced resulting in enhanced coagulation and larger particles. It can also be observed that the predicted particle size distribution is in good agreement with the published experimental data.

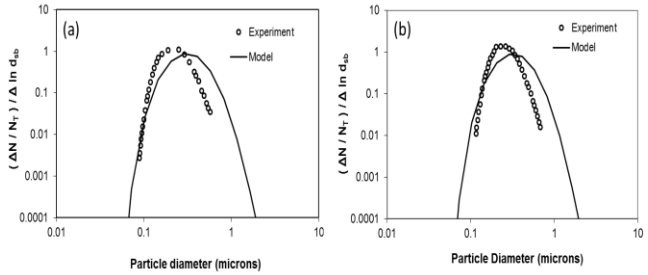


Figure 3: Comparison of predicted aggregate size distribution with experimental data [6] at a furnace temperature of (a) 1600 K and (b) 1723 K ; $TiCl_4$ concentration: $1.16\times10^{-5}$ mol/l in both cases.

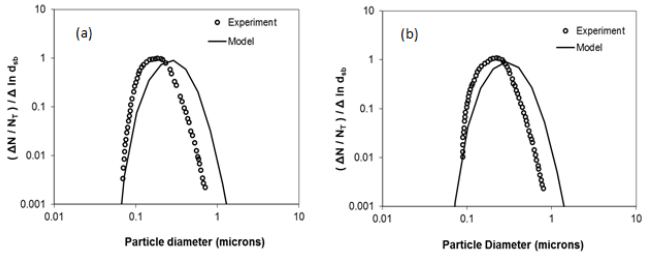


Figure 4: Comparison of predicted aggregate size distribution with experimental data [6] at a furnace temperature of 1400 K and $TiCl_4$ concentration (a) $1.16\times10^{-5}$ mol/l ; (b) $1.56\times10^{-5}$ mol/l.

## 3 CONCLUSIONS

A coupled flame dynamics – 1D PBM model has been developed for synthesis of nanoparticles in a furnace reactor. The flame dynamics was simulated using the commercial computational fluid dynamics software CFX

and the particle population dynamics was represented using a 1D sectional population balance model that predicts the evolution of particle size distribution. The model was tested with published experimental data for synthesis of titania nanoparticles in a furnace reactor. The model predictions for axial gas temperature profiles and aggregate size distributions are in close agreement with published experimental data. The effect of furnace set temperature and initial $TiCl_4$ concentration on the aggregate size distribution within the reactor has been studied and it has been observed that the increase of both these parameters increases the aggregate size.

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