2-D nodal model with turbulent effects for the synthesis of Si nano-particles in RF thermal plasmas

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Abstract: Simulations of the synthesis of Si nano-particles have been carried out using a nodal method and a moment method in a 2-D geometry including a RF plasma apparatus and a reaction chamber. Turbulent effects on the nano-particle formation process have been considered and the model used for their evaluation is presented. Comparisons between the results obtained with the nodal method and the moment method have been reported and the extended amount of obtainable information that characterizes the present model has been underlined.

Keywords: thermal plasmas, nano-particles, turbulence

1. Introduction

Synthesis of nano-particles by means of inductively coupled plasma torches (ICPT) is a material process of large technological interest [1], with numerous parameters concurring in its optimization; hence, the development of numerical models for the prediction of thermal and magneto fluid dynamics fields [2], trajectories and thermal history of precursor powder [3] as well as nano-particle formation and growth is necessary for the up-scaling of these procedures.

Nano-particle formation in plasma systems has been modelled extensively in the past [4, 5], mainly using two different numerical approaches to solve the aerosol general dynamic equation (GDE): the moment method and the discrete-sectional method. While the moment method can save computational time, it often carries the assumption of a uni-modal log-normal profile of the particle size distribution (PSD) for mathematical closure [6]. Contrarily, in discrete-sectional method [7] no assumptions are made on the PSD, but the resulting computational expense is higher than with the moment method. A third model, nodal method [8] in which the treatment of condensation is modified from the idea of nodal splitting scheme [9], has been used and the coupling between the vapour phase and the particle discrete regime by a nucleation source term. Due to the high computational effort implied by the nodal method, a 1-D simplified domain has been adopted in all previous studies. Considering the strong multidimensional behaviour of thermal and fluid dynamic fields generated by an ICPT inside the reaction chamber, the 1-D assumption should be avoided in order to obtain realistic predictions.

Moreover, despite the strong turbulent behaviour typically displayed by the thermo fluid dynamic fields inside the reaction chamber, no computational model currently accounts for the influence of turbulence on the particle synthesis process.

In this work, a 2-D nodal model for the analysis of nano-powder synthesis, taking into account convection, diffusion and the effects of turbulence on particle motion and growth has been used to model the particle production process in a RF plasma source with a reaction chamber, using Si as precursor material. The results obtained with the proposed model have been compared to the ones obtainable using a more widespread nucleation-coupled moment method. Satisfying agreement between the results has been found, although a bi-normal behaviour of the PSD has been evidenced with nodal modelling, especially in recirculation areas. Furthermore, turbulence has been found to limit the growth of the particles, contrasting the effects induced by recirculation.

2. Computational domain and boundary conditions

Plasma synthesis of nano-particles has been investigated in a 2-D domain including a PL-35 Tekna plasma source and an axysimmetric reaction chamber, schematically shown in Figure 1.

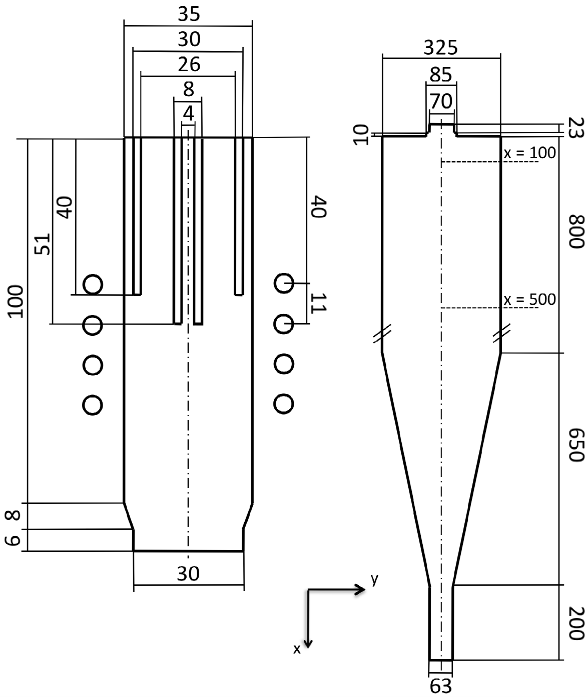

Figure 1. *Detail of the computational domain (dimensions in mm).*

Working gases are supplied through three inlets: carrier gas from the axially movable probe tip, primary gas from the gap between the probe and the quartz tube and sheath gas from the inlet between the quartz and ceramic tubes; mass flow rates are 4 slpm of *Ar*, 12 slpm of *Ar* and 60 slpm of *Ar* + 6 slpm of H_2 for carrier gas, central gas and sheath gas respectively.

A no-slip boundary condition is assumed on internal walls and a 300 K temperature has been applied on the external walls of the torch and the internal walls of the chamber. The torch and the reaction chamber operate at 400 kPa and the power coupled to the plasma is set at 5 kW.

Si precursor particles with a 4 μm diameter are injected through the carrier gas inlet with a mass flow rate of 3 g/min; complete precursor evaporation is obtained.

3. Modelling approach for nano-particle synthesis with turbulent effects

Preliminary studies of plasma thermo fluid dynamic fields and precursor behaviour have been carried out implementing the same models used in [2] and in [10], respectively. Results for temperature and stream function fields are reported in Figure 2.

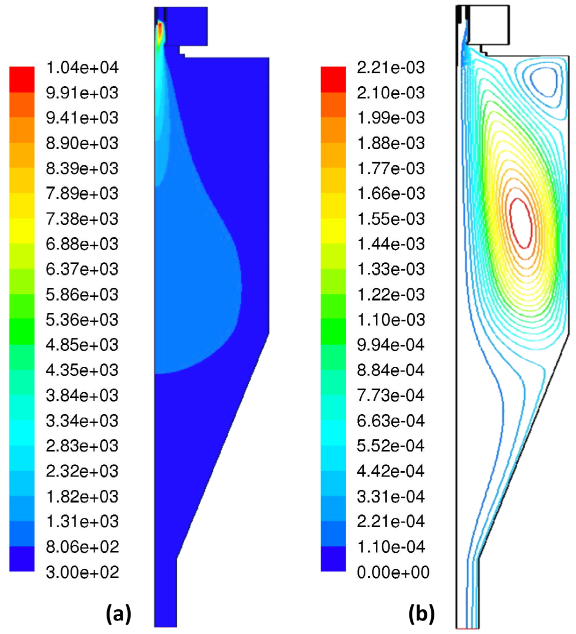

Figure 2. *Detail of the thermo fluid dynamic fields inside the reaction chamber: temperature (K) field (a); streamlines (n).*

Using the results from preliminary simulations as inputs, nano-particle synthesis has been modelled using two different approaches: the nodal model proposed by Shigeta and Watanabe [8] and the moment model presented by Pratsinis and Kim [6]. Both methods have been implemented in the FLUENT© environment in a 2-D geometry.

Turbulent effects in nano-powder production are usually taken into account only for determining precursor vapour behaviour, adding a turbulent term to the classical laminar diffusion coefficient. This treatment is here extended to account for turbulent effects on nano-particle motion, resulting in a particle diffusion coefficient D_p^{tot} defined as:

$$D_p^{tot} = D_p^L + D_p^T$$

where D_p^L is the usual laminar diffusion coefficient and the turbulent diffusion coefficient D_p^T is expressed by:

$$D_p^T = \frac{\mu_T}{Sc\rho}$$

where μ_T and ρ are the turbulent viscosity and the density of the fluid, respectively, while Sc is the Schmidt number.

As eddies dimensions inside the chamber are much larger than particle diameter, it is safe to assume that processes involving interaction between different particles or between particle and vapor are completely unaffected by turbulence. Indeed, these particles are influenced in the same way by the surrounding vortex, leaving the interaction process unbiased.

4. Modelling results

Some selected results concerning particle mean diameter inside the reaction chamber and particle cumulative distribution at reaction chamber outlet on the axis are reported in Figure 3 and Figure 4, respectively. Although the nodal model and the moment model provide a similar tendency, the difference between them should also be underlined.

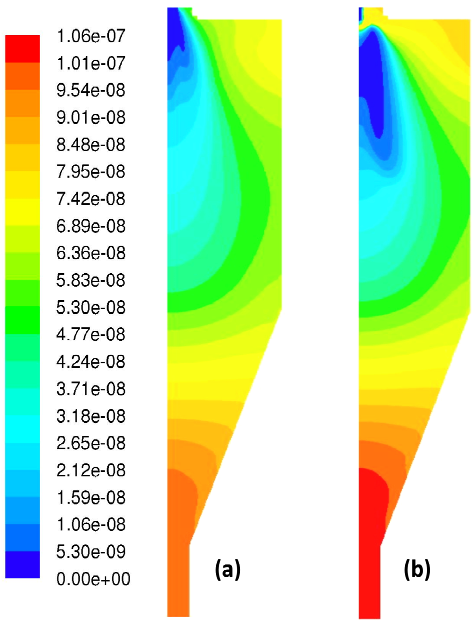

Figure 3. *Detail of the particle mean diameter (nm) inside the reaction chamber: moment model (a) and nodal model (b).*

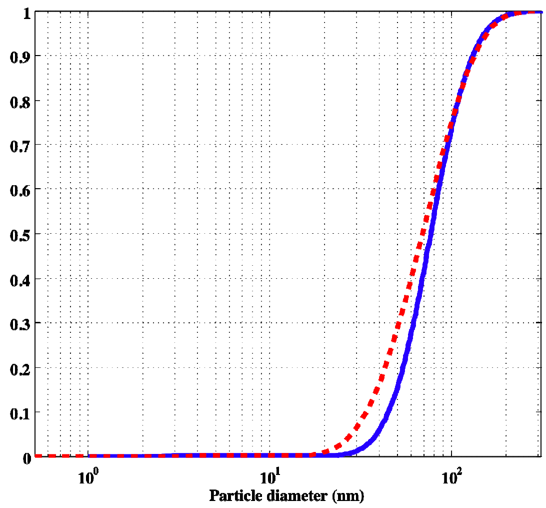

Figure 3. *Detail of the cumulative distribution as a function of the particle diameter (nm) at reaction chamber outlet on the axis: moment model (continuous line) and nodal model (broken line).*

The nodal model is capable to describe the transport characteristics depending on each particle size along with the multi-modal size distribution. On the other hand, the moment model treats an averaged particle transports because of its simplified size distribution. These facts result in different growth behaviours and different particle dispersions.

In Figure 5 and Figure 6, the recirculation induced bi-modal PSD distribution resulting from the nodal model is presented for two different axial quotes. In analogy, the PSD distribution resulting from the nodal model without considering turbulent effects is reported in Figure 7 and Figure 8. Clearly, avoiding

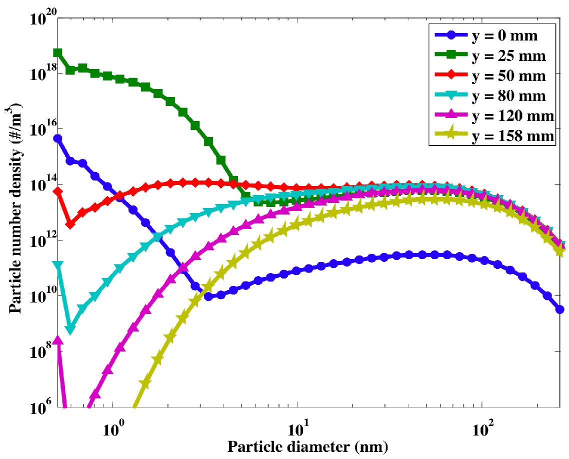

Figure 4. *Detail of the nodal model PSD (1/m^3) inside the reaction chamber at x = 100 mm and various radial (y-axis) positions, considering turbulent effects.*

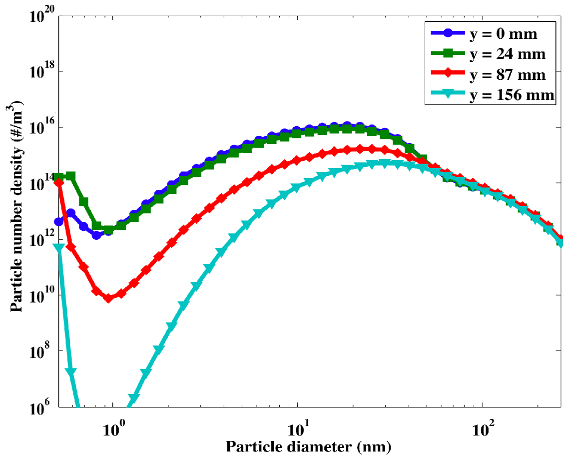

Figure 5. *Detail of the nodal model PSD (1/m^3) inside the reaction chamber at x = 500 mm and various radial (y-axis) positions considering turbulent effects.*

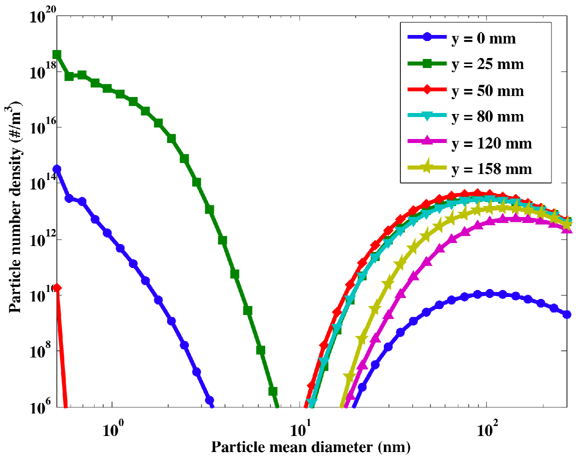

Figure 6. *Detail of the nodal model PSD (1/m^3) inside the reaction chamber at x = 100 mm and various radial (y-axis) positions without considering turbulent effects.*

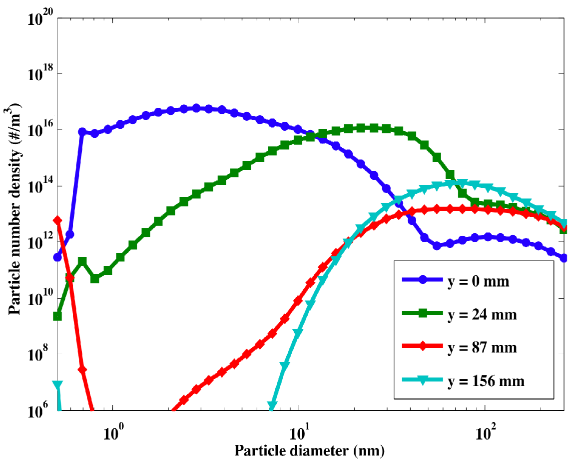

Figure 7. Detail of the nodal model PSD (1/m^3) inside the reaction chamber at x = 500 mm and various radial (y-axis) positions without considering turbulent effects.

the analysis of turbulent effects leads to a more pronounced bi-modal behaviour of the PSD, comparing the same axial quotes; indeed, turbulence acts towards dampening the effect of recirculation, reducing the residence time of particles inside the vortices, thus allowing their distribution along a more uniform profile.

5. Conclusions

2-D simulations of nano-particle synthesis by means of an RF plasma source have been carried out in the framework of the commercial CFD code FLUENT, using both the nodal model and the moment model to describe the processes of nucleation and growth. Effects of turbulence on particle diffusion have been included in the adopted models. Simulation results from both the implemented methods have been displayed to show their similarities and differences. The potential of the nodal model has been underlined with a bi-modal profile in the PSD in recirculation areas. Turbulence enhanced diffusion has been shown to lead to more uniform PSD profiles.

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