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Simulation study of production of fine ceramic powders in a cyclone reactor

Giovanni Manenti, Maurizio Masi*

Dipartimento di Chimica, Materiali e Ingegneria Chimica "Giulio Natta", Politecnico di Milano, Via Mancinelli 7, 20131 Milano, Italy

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ABSTRACT

A numerical simulation study of production of fine ceramic powders in an innovative vapor-phase aerosol reactor is described. Arrangement is typical of reverse-flow cyclone equipment; no similar device is present in current scientific literature and industrial technology. The cyclone reactor has a potential technological application as it realizes process intensification by two simultaneous operating advantages: (i) curly flow reduces recirculation of as-synthesized particles towards flame region, and (ii) cyclone arrangement segregates large particles. As a result, ceramic powders with narrower particle size distribution can be produced with regard to traditional equipment. The study is based on the re-modeling of an existing industrial reactor for production of fine TiO_2 according to a cyclone configuration; particle size distributions from simulation and plant are compared.

1. Introduction

Fine ceramic powders, like titania and zirconia, are high-engineered, high-quality inorganic materials with specific functional properties [1,2]. They are massively used as pigments, catalyst sub-layers and protective coatings [3] and, more recently, as special materials in a broad range of advanced technological applications. For massive industrial productions, vapor-phase synthesis is applied: vapor-phase precursors are fed into aerosol reactors in un-premixed conditions, where gas-to-particle conversion occurs preferably via chemical reaction.

Vapor-phase aerosol reactors intrinsically deviate from ideal behavior for un-premixing of reactants and fast chemical kinetics. Deviation is remarkable when reactors operate under turbulent conditions, which are typical of high-load productions; accordingly, aerosol dynamics results strongly coupled to velocity field [4]. Nozzles configuration and turbulent fluid-mechanics may generate back-flows and vortices where particles are re-circulated and exposed to abnormal growth, which leads to a final product of lower quality. Therefore, from a reactor engineering standpoint, analysis of particle residence time distributions (RTD) becomes essential for understanding and controlling particle evolution [5,6]. As shown in a previous work [7], aerosol reactors designed with traditional axial-symmetric and jet-opposed nozzles present back-mixing, whereas tangential nozzles configurations can reduce it and, consequently, allow synthesizing a product of narrower particle size distribution (PSD). Following such results, a new design for vapor-phase aerosol reactors is proposed in this study. Configuration is similar to process cyclones used for gas–solid segregation, as sketched in Fig. 1; the device simultaneously works as traditional aerosol reactor where particles are nucleated and grown and as separation equipment where the largest particles are removed.

This innovative reactor can reduce the average RTD and improve final quality of fine ceramic powder as (i) back-flows and dead zones are significantly reduced and (ii) right-hand-side tail of the PSD can be cut by segregating the largest particles. As a consequence, use of bag filters or similar expensive segregating devices at the reactor outlet becomes less critical, and the vapor-phase process results intensified. Cyclone reactors have known for years in process industry and several patents have been deposited for applications like volatile organic combustion (VOC) and gas–gas reactions catalyzed by solid particles [8–10]. Reacting and multi-phase systems in cyclones have been discussed in scientific literature also; studies on pyrolysis of biomass [11] and acid gas removal [12,13] represent interesting applications. However, no cyclone reactors have been so far applied to vapor-phase production of fine ceramic powders.

The simulation case study described in this work refers to an industrial reactor for synthesizing TiO_2 micro-powder via hydrolysis of $TiCl_4$, which is a well-run chemical nucleation process [14,15]; relevant numerical model for aerosol synthesis has been discussed in detail and validated elsewhere [7]. The industrial reactor has been re-modeled with a cyclone arrangement, keeping all the operating parameters as per the original, by means of a commercial computational fluid-dynamics (CFD) code. Computational results, under modeling assumptions, show that standard deviation of PSD of as-synthesized powder can be reduced and therefore quality of final product can be improved by use of a cyclone reactor. This work represents a first attempt to investigate the feasibility of an aerosol

* Corresponding author. Tel.: +39 02 2399 3131; fax: +39 02 2399 3180.
E-mail address: maurizio.masi@polimi.it (M. Masi).

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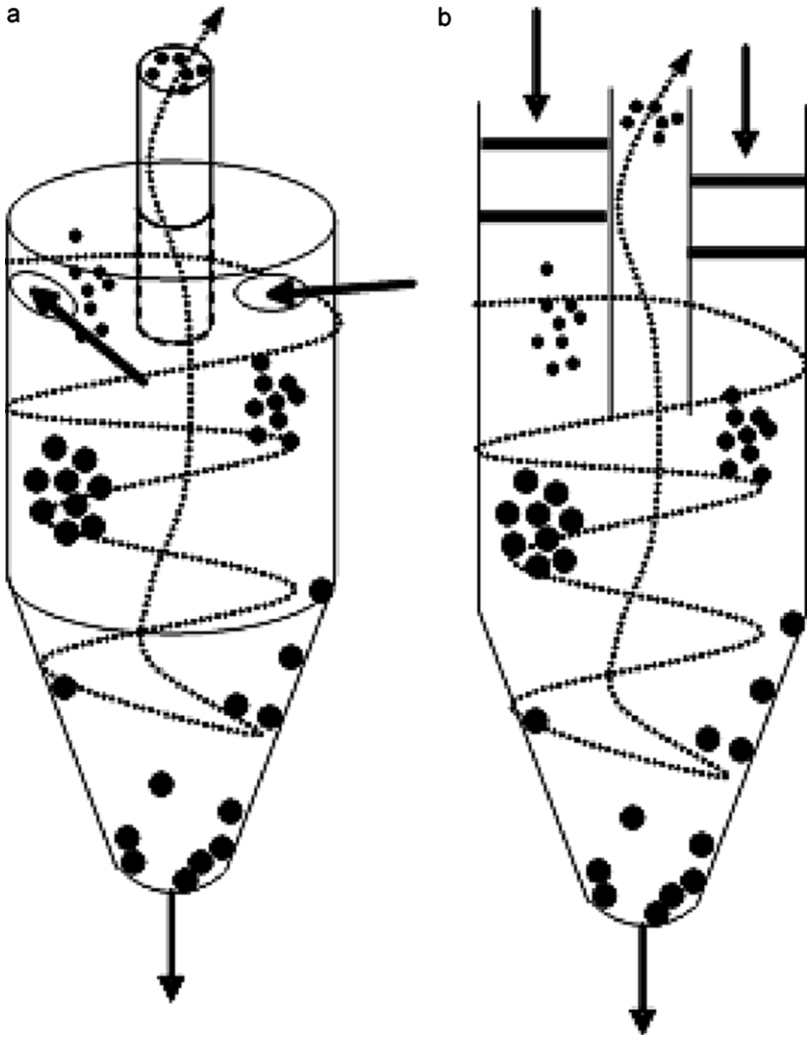

Fig. 1. Conceptual arrangements for an aerosol cyclone reactor: tangential inlets (a) and axial inlets (b).

cyclone reactor, thus quantitative trends are searched rather than final results.

2. Reactor model

2.1. Aerosol dynamics

Chemical nucleation of TiO_2 particles can be described by following one-step hydrolysis reaction:

$$TiCl_4 + 2H_2O \rightarrow TiO_2 + 4HCl \quad (1)$$

Assuming negligible activation energy due to fast kinetics and high temperatures, chemical kinetic rate, R, is a function of species concentrations and fluid temperature, C_i and T, and has been found to be [16]:

$$R = k\sqrt{T}C_{TiCl_4}C_{H_2O} \quad (2)$$

The frequency factor, $k = 2.4 \times 10^{10}\ m^3\ \#^{-1}\ K^{-1/2}\ s^{-1}$, has been obtained via a quantum mechanical method based on density functional theory (DFT) [17,18].

Mass, momentum, energy and chemical species conservation equations for fluid are expressed by Eq. (3). Since Peclet number is $\gg 1$, contribution of diffusive terms is considered negligible; this leads to a computational problem which is less time consuming. Reactor walls are kept at the bulk temperature.

$$\begin{aligned} &\nabla\cdot(\rho\mathbf{u}) = 0 \\ &\nabla\cdot(\rho\mathbf{u}\cdot\mathbf{u}) = \nabla\cdot\Sigma \\ &\nabla\cdot(\rho\mathbf{u}H) = k\sqrt{T}C_{TiCl_4}C_{H_2O}H_R \\ &\nabla\cdot(\rho\mathbf{u}C_i) = k\sqrt{T}C_{TiCl_4}C_{H_2O} \end{aligned} \quad (3)$$

Fine ceramic powder produced in the industrial reactor under study presents a log-normal PSD; this is in accordance with experimental observations on processes for synthesizing metal-oxide particles [19,20]. Consequently, shape of the distribution can be assumed a priori and method of moments can be applied for describing the particle population balance. Only nucleation and coagulation phenomena are accounted for, assuming negligible breakage and other growing contributes. As TiO_2 monomers are thermodynamically stable even at high temperatures [16,21], particle nucleation rate corresponds to chemical kinetic rate; at simulated operating conditions, TiO_2 is solid and precursors and carrier fluid do not condense. Spherical particles are considered. Accordingly, aerosol dynamics is effectively described by three conservative equations which depending variables, usually called "moments" of the PSD, are (i) the particles concentration, (ii) the particles volume and (iii) the spread of the particles distribution; they are denoted by M_0, M_1 and M_2 respectively. For stationary