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Control of particulate processes: Recent results and future challenges ☆

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Abstract

This paper provides a discussion of the existing results on control of particulate processes using population balance models and presents an overview of future research directions in this field in the context of chemical, materials and biological process systems.

Keywords: Nonlinear control; Particle size distribution; Model reduction; Crystallization processes; Aerosol reactors; Spray coating processes

1. Introduction

Particulate processes are prevalent in a number of process industries including agricultural, chemical, materials, food, minerals, and pharmaceuticals. By some estimates, 60% of the products in the chemical industry are manufactured as particulates with an additional 20% using powders as ingredients. Representative industrial particulate processes include the crystallization of proteins for pharmaceutical applications, the emulsion polymerization for the production of latex, the fluidized bed production of solar-grade silicon particles through thermal decomposition of silane gas, the aerosol synthesis of titania powder used in the production of white pigments, and the thermal spray processing of nanostructured thermal barrier and wear resistant coatings. One of the key attributes of particulate processes is the co-presence of a continuous phase and a dispersed phase, which leads to the occurrence of physico-chemical phenomena such as particle nucleation, growth, coagulation, breakage, melting and solidification. These phenomena are absent in homogeneous processes and lead to physical and chemical properties of the particulate product such as particle size, shape, morphology, porosity and molecular weight, which in turn, determine the physico-chemical and mechanical properties of the final products. For example, a nearly mono-disperse powder size distribution around 0.2–0.3 μm is required for the titania pigments to obtain the maximum opacity or hiding power [14]. As another example, the size, velocity, temperature and degree of melting of powder particles at the point of impact on the substrate in thermal spray coating processes have a direct effect on the microstructure of the coating and its mechanical and thermal properties [19]. Finally, in crystallization processes, the shape of the crystal size distribution is an important quality index which strongly affects crystal function and downstream processing such as filtration, centrifugation, and milling [58]. In all of these instances, the particle size distribution (PSD) provides the critical link between the product quality indices and the process operating variables; thus, the ability to effectively control the shape of the PSD is essential for regulating the end product quality in these processes.

Population balances have provided a natural framework for the mathematical modeling of PSDs (see, for example, the tutorial article [22] and the review article [55]), and have been successfully used to describe PSDs in emulsion polymerization reactors (e.g., [10]), crystallizers (e.g., [58]), aerosol reactors (e.g., [14,80]) and cell cultures (e.g., [9]). Application of population balances to particulate processes typically leads to systems of nonlinear partial integro-differential equations that describe the rate of change of the PSD. The population balance models are

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intrinsically coupled with the material, momentum and energy balances that describe the rate of change of the state variables of the continuous phase (these are usually systems of nonlinear differential equations, which include integrals over the entire particle-size spectrum), leading to complete particulate process models. The nonlinear and distributed nature of population balances has motivated extensive research on the development of efficient numerical methods for the accurate computation of their solution. Examples of solution methods for continuous population balances include the method of self-preserving distributions [14], the method of weighted residuals (e.g., [55,15]), the sectional method (e.g., [16,30]), and discretization via fixed/moving pivot techniques ([28,29]) and finite-differences/elements [44,45], while methods for the solution of discretized population balances have been proposed by [21] and [20]. An excellent review of results in this area can be found in [55,9]. The ability to accurately solve population balance models has also motivated numerous research studies on the dynamics of particulate processes. These studies confirmed the existence of a wide range of complex static and dynamic bifurcation phenomena including multiple steady-states and sustained oscillations (see, for example, [31,23,59,60] for a theoretical analysis of oscillatory behavior in crystallizers and emulsion polymerization reactors, respectively), which had been previously observed in experimental studies [56].

In spite of the rich literature on population balance modeling, numerical solution, and dynamical analysis of particulate processes, up to about ten years ago, research on model-based control of particulate processes had been very limited. Specifically, early research efforts had mainly focused on the understanding of fundamental control–theoretic properties (controllability and observability) of population balance models [62] and the application of conventional control schemes (such as proportional–integral and proportional–integral–derivative control, self-tuning control) to crystallizers and emulsion polymerization processes (see, for example, [63,61,10] and the references therein). The main difficulty in synthesizing nonlinear model-based feedback controllers for particulate processes is the distributed parameter nature of the population balance models which does not allow their direct use for the synthesis of low-order (and therefore, practically implementable) nonlinear output feedback controllers. Furthermore, a direct application of the aforementioned solution methods to derive finite dimensional approximations of the population balance models typically leads to ordinary differential equation (ODE) systems of very high order, which are inappropriate for the synthesis of low-order controllers. This limitation had been the bottleneck for model-based synthesis and real-time implementation of nonlinear feedback controllers on particulate processes.

2. Recent developments

2.1. Control theory for particulate processes

Motivated by the lack of population balance-based control methods for particulate processes and the need to achieve tight size distribution control in many particulate processes, we developed, over the last ten years, general nonlinear, robust and predictive control methods for particulate processes based on population balance models [8,5,24,6,12,26,7,64,66]. The development of these control methods has been based on the property that the dominant dynamic behavior of many particulate process models is low-dimensional. Examples of this fundamental property include the occurrence of oscillatory behavior in continuous crystallizers and the ability to capture the long-term behavior of aerosol systems with self-similar solutions. Taking advantage of this property, we proposed a model reduction procedure, based on a combination of the method of weighted residuals and the concept of approximate inertial manifold, which leads to the construction of low-order ODE systems that accurately reproduce the dominant dynamics of broad classes of particulate process models. These ODE systems were subsequently used for the synthesis of nonlinear feedback controllers that enforce exponential stability in the closed-loop system. Within the developed framework, the infinite dimensional closed-loop system stability, performance and robustness properties were precisely characterized in terms of the accuracy of the approximation of the low-dimensional models. Furthermore, controller designs were proposed that deal directly with the key practical issues of uncertainty in model parameters, unmodeled actuator/sensor dynamics and constraints in the capacity of control actuators and the magnitude of the process state variables. It is also important to note that owing to the low-dimensional structure of the controllers, the computation of the control action involves the solution of a small set of ODEs, and thus, the developed controllers can be readily implemented in real-time with reasonable computing power, thereby resolving the main issue on model-based control of particulate processes. In addition to theoretical developments, we also successfully demonstrated the application of the proposed methods to size distribution control in continuous and batch crystallization, and aerosol and thermal spray processes and documented their effectiveness and advantages with respect to conventional control methods. In the following subsections, we discuss the main results and insights of these application studies carried out in the context of our research work; the reader may refer to [9,11,4] for recent reviews of results on simulation and control of particulate processes.

2.2. Control of crystallization processes

Crystallization is a particulate process which is widely used in industry for the production of many products including fertilizers, proteins, and pesticides. The fact that the shape of the crystal size distribution (CSD) influences significantly the necessary liquid–solid separation, as well as the properties of the product, implies that crystallization requires a population balance in order to be accurately described, analyzed, and controlled. We applied the developed methods for feedback control of particulate processes to both continuous and batch crystallization processes. Continuous crystallizers typically exhibit highly oscillatory behavior which motivates the use of feedback control to ensure stable operation. In the context of continuous crystallization control, we first constructed low-dimensional approximations of the population balance [5]. These approximate models were subsequently used for the design of nonlinear feedback controllers which utilize measurements of the total crystal concentration and manipulate the solute feed concentration to achieve closed-loop stability and attain a crystal size distribution with desired characteristics. We successfully validated the performance of the nonlinear controllers through application to a detailed population balance model and demonstrated their superiority over linear control schemes [5]. Further, we successfully developed and implemented on population balance models nonlinear feedback designs which deal explicitly with parametric model uncertainty, unmodeled actuator/sensor dynamics [6] and manipulated input constraints [12]. In addition to continuous crystallization, we also applied the developed control methods to batch crystallization systems. In this case, the control objective was to achieve CSD with desired characteristics subject to both manipulated input and product quality constraints by manipulating the crystallizer temperature. An optimization-based predictive control strategy that incorporates these constraints explicitly in the controller design was formulated and applied to a seeded batch cooling crystallizer [64] and a protein crystallizer [66]. We demonstrated the achievement of CSDs of desired shape at the end of the batch, improvement over standard cooling strategies, and robustness with respect to modeling errors.

2.3. Control of aerosol reactors

Aerosol processes are increasingly being used for the large scale production of nano-and micron-sized particles. These processes have largely replaced other processes which involve multiple steps of wet chemistry, due to the direct gas phase chemical reaction of precursor vapor and the ease of separation of the particulate products from the gas. Aerosol products, such as TiO_2, B_4C , find widespread use as pigments, reinforcing agents, ceramic powders, optical fibers, carbon blacks and semiconductor materials. We developed [24] a nonlinear feedback control method for spatially-inhomogeneous aerosol processes for which the manipulated inputs, the control objectives and the measurements were distributed in space. Our starting point was a general nonlinear partial integro-differential equation model describing aerosol processes with simultaneous chemical reaction, nucleation, condensation, coagulation and convective transport. Then, under the assumption of lognormal aerosol size distribution, the method of moments was employed to reduce the original model into a set of first-order hyperbolic PDEs which accurately described the spatio-temporal evolution of the three leading moments needed to exactly characterize the aerosol size distribution. This hyperbolic PDE system was 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). We applied [25] our control method to an aerosol flow reactor used to produce titania powder by gas phase oxidation of titanium tetrachloride. The controller manipulates the temperature of the reactor wall to achieve an aerosol size distribution in the outlet of the reactor with desired geometric average particle diameter. The performance and robustness of the nonlinear controller was successfully tested through computer simulations and was found to outperform linear control techniques.

2.4. Control of thermal spray coating processes

The high velocity oxygen fuel (HVOF) thermal spray process is a particulate deposition process widely used in aerospace and automobile industry to deposit coatings on a substrate in order to extend product life, increase performance and reduce production and maintenance costs. While the analysis and control of the HVOF process has been largely empirical, our research led to the development of a computational methodology for controlling the coating microstructure which determines the coating mechanical and physical properties by manipulating macro-scale operating conditions such as gas feed flow rates and spray distance. The major challenge on this problem was the development of multiscale models linking the macroscopic scale process behavior (i.e., gas dynamics and particle inflight behavior) and the microscopic scale process characteristics (evolution of coating microstructure), and the integration of models, measurements, and control theory to develop measurement/model-based control strategies. Specifically, we developed a computational framework for the HVOF thermal spray processing of nanostructured coatings [32,36,65,33,37,38,34,35]. The multiscale process model encompasses gas dynamics of the supersonic reacting flow, evolution of particle velocity, temperature and molten state during flight, and stochastic growth of coating microstructure. Parametric analysis based on the multiscale model pointed out the coating microstructure is highly dependent on particle velocity, temperature and molten state at impact on substrate, which can be almost independently adjusted by pressure in the combustion chamber and fuel/oxygen ratio. A model-based control scheme was developed based on the gas phase measurement and the estimation of particle properties through the dynamic particle inflight model and was designed to control the particle velocity and melting ratio at impact by adjusting the flow rate of cooling air, oxygen and fuel, through which the pressure and fuel/oxygen can be independently adjusted. The multivariable feedback control system was applied to a detailed mathematical model of the process and the closed-loop simulations showed that the proposed controller was effective in set-point tracking and also robust with respect to disturbances in the processing environment, such as spray distance and particle injection velocity, and variations in powder size distribution.

3. Future research challenges

While significant work has been done on control theory for particulate processes over the last ten years, there is a number of emerging applications within the areas of nanotechnology, advanced materials processing and energy storage and conversion where control of distributed particle characteristics such as size, morphology and composition could be used as tools for optimizing the performance and robustness of key processes.

3.1. Control of multiple distributed nanoparticle properties during synthesis

Significant interest has been generated recently in the field of nanoparticle synthesis and processing which has potential applications in the manufacture of catalysts, coatings, fillers, sensors, membranes and ceramics. The nanostructured materials fabricated by the processing of nanoparticles (characterized by a grain size less than 100 nm) exhibit superior qualities to conventional counterparts due to larger surface area to volume ratio, larger grain boundaries/interfaces etc. For example, nanostructured tungsten oxide coatings prepared by aerosol-assisted chemical vapor deposition (CVD) demonstrate much higher photocatalytic activity than those prepared by conventional CVD processes. Furthermore, nanostructured tin dioxide sensors prepared by flame spray pyrolysis with particle size less than 10 nm show higher sensitivity and selectivity than conventional gas sensors [50]. Finally, nanostructured solid oxide fuel cells (SOFCs) prepared by laser reactive deposition have lower operating temperature and higher fuel utilization than common SOFCs.

In these and many other processes nanoparticles have multiple distributed characteristics. For most nanoparticles the smallest physical scale is the crystallite size. Often the crystallite size defines the primary particle size. This length scale has direct implications on desired nanoparticle properties such as surface chemistry (catalysis), interaction with electromagnetic waves (quantum dots, fillers) and electron density (sensors). Many applications require a close control of this distributed particle length scale (PSD) in order to achieve the highest performance. In addition, most nanoparticles agglomerate during their synthesis or processing. Such agglomerates are formed due to the collisions of compact (often spheroidal) primary particles or agglomerates themselves at temperatures sufficiently low to avoid complete coalescence before the next collision occurs [2]. The resulting contacts between the primary particles may be of chemical nature (covalent and ionic bonding) resulting from neck formation between the nanoparticles. In this case the primary particles form hard agglomerates characterized by high particle binding energies [75]. The fractal like nature of agglomerates can be mathematically expressed through a statistical scaling law. The degree of agglomeration and final aggregate size distribution (ASD) is determined by simultaneously gas phase and surface reactions, coagulation and sintering. The control of the ASD presents a major challenge. To describe the evolution of the ASD in these processes, a general aerosol dynamic equation might be necessary and approaches such as the ones proposed in [24,25] might be followed for the particle-size distribution control in these processes. The ASD influences for example packing density (catalysts, ceramics), dispersion (filler), and electronic pathways (sensors).

Most of today's nanoparticles are multicomponent systems where the spatial distribution of mixing is a key parameter. The required mixing length scale depends on the material systems and applications. For example, the mixing of CeO_2 and ZrO_2 in a solute solution (mixing within the crystal) resulted in very stable and active oxygen exchange catalysts in contrast to individual nanoparticles of these components [67]. In comparison the mixing in the aggregate length scale of individual Al_2O_3 and monoclinic $BaCO_3$ nanoparticles exhibited excellent NO_x storage activity in contrast to a catalyst with both components in the primary particle [69]. Today, key control parameters such as precursor initial volume fraction, maximum temperature, residence time, cooling rate, reaction and restructuring kinetics have been identified. Future research and development will focus on multicomponent/multifunctional nanomaterials. This will increase the complexity of these processes and the demands for their control. For example, flame spray pyrolysis further broadens the spectrum of flame made powders and their use in various applications as there are more liquid than gaseous precursors available [46]. Applications of such functional nanoparticles include: dental fillers [47], UV-filters [51] and catalysts [74]. The introduction of a third phase in the process chain, the liquid precursor droplets which evaporate and react, poses an additional complexity for control. Furthermore, the mass loading and the released energy of the liquid precursor droplets result in momentum and heat transfer with the surrounding gas and require therefore the control of all phases: suspending media, precursor droplets and product particles. In the area of nanoparticle synthesis, another process where PSD control could make a difference is the synthesis of quantum dot nanoparticles via liquid phase routes [41]. This approach offers significant advantages over gas phase routes because of the lack of aggregation, which is frequently present when gas-phase routes are followed.

3.2. Control of spray coating processes

Spray pyrolysis is a process in which an aqueous suspension of particulate precursors is atomized and transported in a gas stream to a hot surface where a solid product is formed as a result of the pyrolytic decomposition of the organometallic precursor. It is a cost effective and versatile technique to deposit metal or metal oxide thin films on large substrate areas. Such a process has been used in the glass industry for more than fifty years to produce transparent conductive oxides [52] and solar control metal oxides [73]. It is also used in the fabrication of solar cells [54], SOFC [79] and sensors [17]. However, the quality of the coatings is sensitive to the operating conditions such as substrate temperature, droplet size, solution concentration, carrier gas flow rate and nozzle-to-substrate distance [53,78]. These parameters directly influence the microstructure, crystallinity and morphology of the coatings and the resulting mechanical and physical properties. For example, a too high temperature might cause a high surface roughness and a porous microstructure. A low temperature, however, may lead to coating cracking. If evaporation is much faster than diffusion as a result of rapid heating, hollow particles might be formed. Moreover, a droplet may either land and flatten or skip along the surface, depending on its density, size, velocity and the local flow, thermal and electrostatic fields. Therefore, to improve the coating quality and to maximize the deposition efficiency requires that the key process parameters should be tightly controlled.

In the area of nanostructured coatings processing, it has been recently reported that atomized liquid precursors can be fed to a plasma to make nanostructured ceramic coatings (e.g., [70,71,3]). In contrast to the conventional thermal spray process, the precursor gases are involved in chemical reactions which result in nano-grained particles. Depending on the residence time of the droplets in the high temperature environment, the reaction might occur in the plasma stream, near the surface of the substrate or on the surface, leading to different type of deposits. If incomplete evaporation of the solvent occurs before the droplet hits the substrate, the droplet might splash on the surface and undergo further reaction, resulting in a patchy coating. However, if the powder particles are formed in the plasma, they may result in agglomerates through coagulation, leading to a powdery coating. The best quality coatings are obtained when the reaction occurs near the surface, where particles are formed and directly deposited on the substrate. In order to accomplish this scenario, precise control of the residence time distribution and size of the particles formed on the surface by regulating process parameters such as precursor concentration, plasma temperature and carrier flow rates as well as the spray distance, is needed.

Porous films composed of nanoparticles find applications as catalysts, solar cells, gas sensors, and diesel soot traps. The aerosol route offers several possibilities to fabricate porous layers or coatings by nanoparticle deposition from the gas phase. For example, CO sensors synthesized by the aerosol deposition method have significant advances over the fabrication of conventional particle slurry type coatings [49]. In an ideal film synthesis the film porosity and the film thickness can be controlled independently. Such a control scheme needs to regulate the primary particle size and the agglomerate morphology in order to optimize film growth rate and film structure [48].

3.3. Controlling particulate processes in energy conversion and storage

The production of hydrogen from water using Sol. Energy is considered as one of the best examples of sustainable energy utilization. Specifically, water splitting thermochemical cycle achieved through the metal/metal oxide redox reactions is a promising route for high efficiency and safe hydrogen generation because the separation of H_2/O_2 is circumvented [68]. The central idea is to use several thermodynamically feasible reactions occurring at different temperatures to achieve an overall reaction that is thermodynamically infeasible under normal industrial operating conditions. For example, the water splitting using Zn/ZnO consists of a two-step thermochemical cycle. The first step is the endothermic dissociation of ZnO(s) into Zn(g) and O_2 which occurs at temperatures above 2000 K. The second reaction is the exothermic hydrolysis of Zn at 700 K to form H_2 and ZnO(s). ZnO as a solid separates naturally from the hydrogen gas and is recycled to the first step. Recent efforts have reported a novel hydrogen generation process in which the formation of Zn nanoparticles and the in-situ hydrolysis of hydrogen are integrated in a single tubular aerosol flow reactor [77,76]. A model-based optimization and control of this process is expected to improve the yield of hydrogen and the stability of operation, and to facilitate the commercialization of this process. This requires a dynamic model describing the process which encompasses population balance equations describing the evolution of the particle-size distribution (nucleation, reaction, condensation, coagulation) and the governing equations of the gas phase. However, due to the complex reaction kinetics and short residence time, the model-based optimization and control of this process will be a very challenging task.

3.4. Particle contamination control in advanced material manufacturing processes

While particulate processes have a variety of important applications, particles might be detrimental in some industrial processes. One example is the gas phase nucleation induced particle contamination of wafers in the semiconductor manufacturing industry, which significantly reduces the quality and yield of the computer chips [57]. In the glass coating industry, online chemical vapor deposition provides a cost efficient tool to manufacture a variety of high performance coatings on float glass including, but not limited to energy conserving low-E coatings, self-cleaning coatings and transparent conductive oxide for photovoltaics [18,1,39]. These products are routinely used for architectural, automotive, aircraft, and renewable energy applications. However, due to the high deposition rate, particles generated from gas phase nucleation can deposit and adhere to the transport channels, thereby restricting flow and reducing run length due to the necessity to clean the coating apparatus. Moreover, sodium chloride formed in the coating as a result of the combination of sodium migrated from the soda lime glass and the chlorine generally contained in precursor might lead to intrinsic haze, which may make the coated glass product aesthetically unacceptable [72]. The main challenge in this area is to develop computational models to predict formation, kinetics and transport of particles as a function of the process flow and thermal conditions and to identify the dominant chemical clustering pathways and limiting growth mechanisms. A high-fidelity model will provide guidelines for process and equipment design. Moreover, a development of online diagnosis and control schemes by integration of process models and online measurements will suppress the particle contamination in real-time.

3.5. Control of biological particulate processes

The objective of most biotechnologically relevant applications is to maximize the productivity of entire cell populations instead of that of individual cells. Moreover, the majority of the high-throughput experimental techniques (e.g. transcriptomics, proteomics, metabolomics, x-omics in general) that are currently available to characterize cellular physiology collect measurements from entire cell populations instead of individual cells. Therefore, it is more meaningful to define as the biological system of interest the cell population. However, this shift of focus from the individual cell to the cell population introduces an extra source of complexity: cell populations are heterogeneous systems in the sense that cellular properties are vastly distributed amongst the cells of the population. Cell population balance models [13] represent the only class of mathematical models which take into account cell population heterogeneity. The importance of the latter in making accurate predictions of even average population properties both transiently and asymptotically has been recently documented [40,42,43]. Therefore, successful attempts to rigorously design efficient controllers for controlling cell population dynamics should be based on cell population balance models.

Recent modeling studies [40,42,43] have also shown that for specific single-cell genetic architectures (e.g. lac operon, genetic toggle) steady-state multiplicity exists even at the cell population level. Development of controllers that will stabilize the intermediate unstable steady-state branches using extracellular conditions as manipulated inputs, will offer the opportunity of achieving a wider range of biotechnological objectives especially in cases where artificial genetic networks are used to direct intracellular metabolic fluxes towards desirable metabolic goals. Another more general challenge from the control perspective is to design controllers which can control at the cell population level sources of stochastic noise, typically related to random operator fluctuations or the function of small number of regulatory molecules at the single-cell level [27].

4. Concluding remarks

Control of particulate processes systems is a cross-disciplinary and rapidly growing research area that brings together fundamental modeling, numerical simulation, nonlinear dynamics and control theory. While recent advances have led to systematic methods for the design of easy-to-implement nonlinear feedback controllers for broad classes of particulate processes, the ever-increasing research in nanoparticle synthesis and processing, hydrogen generation, semiconductor manufacturing and cell population modeling provides a large number of new and intellectually-challenging problems. Solutions to these problems could have a significant impact in important application areas such as advanced materials and semiconductor manufacturing, nanotechnology, and biotechnology. The integration of population balance modeling and advanced control into the chemical engineering curriculum will be essential for chemical engineers to play an important role in this rapidly expanding field.

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