A POPULATION BALANCE BASED DESIGN PROBLEM

in a Particle Science and Technology Course

SHERYL H. EHRMAN, PATRICIA CASTELLANOS, VIVEK DWIVEDI, AND R. BERTRUM DIEMER*
University of Maryland • College Park, MD 20742

There is a great need for chemical engineering students to have some exposure to particle technology concepts, since so many products are processed and handled in particle or powder phase. For example, more than half (by volume) of all products sold by DuPont and Dow are in particulate form. Considering raw materials, intermediates, and co-products, the amount of solids handled is actually three to four times the amount sold.[1] In addition, there are increasing opportunities for chemical engineers with some particle technology training in fields outside of the traditional chemical process industry, such as sensing technology development for homeland security, aerosol drug formulation, and device development and nanomaterials processing. Over the past decade, there has been a push to incorporate more particle technology into core chemical engineering coursework, and, as recommended by Nelson, *et al.*,[1] to include elective courses in particle technology. At the University of Maryland, a course designed for senior-level undergraduates and first-year graduate students was created and taught initially in 2002. The course spans "dry" particle technology from aerosol science to powder technology unit operations.

To bridge the gap between fundamental aerosol science and powder technology, a team design problem incorporating population balance modeling and design of particle collection systems was developed and assigned to students in our Particle Science and Technology course. The main motivation for developing this assignment was to provide a realistic open-ended industrial problem for the students to solve that would incorporate population balance modeling and aerosol dynamics to describe evolution of particle size distributions. The problem was developed collaboratively between a representative from industry and a University of Maryland faculty member, with additional input from the graduate student teaching assistant for the course. The problem was assigned twice (spring 2003 and fall 2005), with changes made in the second version based on feedback received from students in the first. Students in the fall 2005 course provided additional feedback.

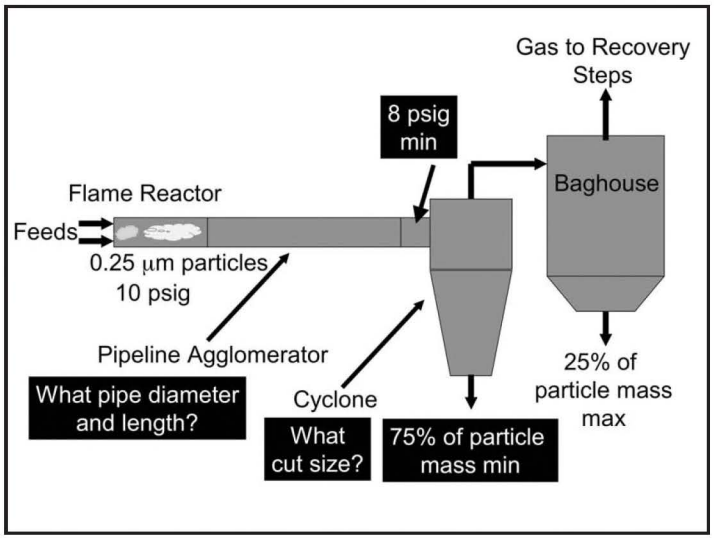

Figure 1. *Schematic of aerosol generation and particle collection system.*

PROBLEM DESCRIPTION

In this problem, students were given a description of a particle synthesis and collection system as shown in Figure 1. The model particles, with properties similar to those of

Sheryl H. Ehrman *is an associate professor in the Department of Chemical and Biomolecular Engineering at the University of Maryland. She earned B.S. and Ph.D. degrees in chemical engineering from the University of California, Santa Barbara, and the University of California Los Angeles, respectively.*

Vivek Dwivedi *is a graduate student in the Department of Chemical and Biomolecular Engineering at the University of Maryland. He has a B.S. and an M.E. in chemical engineering, both from the University of Maryland.*

Patricia Castellanos *is a graduate student in the Department of Chemical and Biomolecular Engineering at the University of Maryland. She has a B.S. in chemistry from the University of Maryland.*

R. Bertrum Diemer *is DuPont's principal division consultant in reaction engineering, thermochemistry, and population balance modeling and an adjunct professor of chemical engineering at the University of Delaware. He received chemical engineering degrees from Lehigh (B.S. '73) and Delaware (M.S. '80, Ph.D. '99) and is a Delaware-licensed P.E.*

* *DuPont Engineering Research and Technology, DuPont Company, Wilmington, DE 19898*

silica particles, were produced in an aerosol reactor. One major simplifying assumption was made: The size distribution of the particles leaving the reactor was assumed to be monodisperse. The particle collection system consisted of a pipeline agglomerator, a cyclone, and a baghouse. The objective of this design was to reduce wear on the baghouse by collecting 75% of the particle mass in the cyclone. Use of the cyclone alone was not feasible for the conditions specified in the problem statement, because standard cyclones typically are not very efficient at collecting sub-micron-diameter particles. Hence the coagulation tube was added prior to the cyclone to promote formation of larger agglomerates that could be collected more efficiently by the cyclone. In both implementations of the problem, in 2003 and 2005, students focused on the pipeline agglomerator and the cyclone in their system design.

Within the pipeline agglomerator, coagulation and breakup were assumed to be the only processes occurring. Assuming steady state, axisymmetric incompressible plug flow, with particle size distribution dynamics varying only along the z axis of the pipeline agglomerator, the simplified form of the aerosol general dynamic equation, expressed in the form of a population balance, is:

$$u_z \frac{\partial n(V)}{\partial z} = \frac{1}{2}\int_0^V \beta(\phi, V-\phi)\, n(\phi)\, n(V-\phi)\, d\phi - n(V)\int_0^\infty \beta(\phi, V)\, n(\phi)\, d\phi + \int_V^\infty \Gamma(\Phi)\, b(V;\Phi)\, n(\Phi)\, d\Phi - \Gamma(\Phi)\, n(V) \quad (1)$$

where u_z is the axial velocity, n is the particle population density function distributed by particle volume in units of number concentration, z is the axial position, V is particle volume, Γ is the breakage rate kernel, Φ is the parent particle volume in breakage, b is the breakage daughter distribution, and β is the coagulation rate kernel. The left-hand side describes the evolution of the volume distribution as a function of the distance down the axis of the pipeline agglomerator. The first and second terms on the right-hand side represent changes in the size distribution because of agglomeration, and the last two terms describe changes to the size distribution resulting from breakup.

Under the conditions specified in the problem statement, Brownian diffusive transport of particles in the z direction was assumed to be negligible. Collisions were assumed to be 100% efficient, with no influence from long-range forces. Coagulation was assumed to result in soft agglomerates, *i.e.*, no sintering of the particles occurred. The coagulation rate kernel was represented as the sum of the Brownian coagulation and the Saffman-Turner rate kernels as:

$$\beta(\phi, V-\phi) = \frac{2kT}{3\mu}\left[2 + \left(\frac{\phi}{V-\phi}\right)^{1/3} + \left(\frac{V-\phi}{\phi}\right)^{1/3}\right] + 0.31\sqrt{\frac{\varepsilon}{\nu}}\left[V + 3\phi^{1/3}(V-\phi)^{2/3} + 3\phi^{2/3}(V-\phi)^{1/3}\right] \quad (2)$$

where k is the Boltzmann constant, T the temperature, μ is the fluid viscosity, ν is the kinematic viscosity, calculated by dividing the fluid viscosity by the density of the total flowing stream (fluid plus particles), and ε is the energy dissipation per unit mass. Breakage was assumed to result in two equal-sized daughters, and this assumption greatly simplified the sectional approach. The breakage kernel is given as:

$$\Gamma(V) = \Gamma_0 \left(\frac{\varepsilon}{\nu}\right)^{3/2} V^{1/3} \quad (3)$$

The daughter distribution is given by:

$$b(V;\Phi) = 2\delta\left(V - \frac{\Phi}{2}\right) \quad (4)$$

where delta = is the Dirac delta function, and V and Φ are defined as described as above.

In discrete form, the population balance Eq. (1) can be expressed in terms of summations

$$u_z \frac{dn_i}{dz} = \frac{1}{2}\sum_{j=1}^{i-1} \beta_{j,i-j} n_j n_{i-j} - n_i \sum_{j=1}^{\infty} \beta_{i,j} n_j + \Gamma_{2i+1} n_{2i+1} + 2\Gamma_{2i} n_{2i} - \Gamma_{2i-1} n_{2i-1} - \Gamma_i n_i \quad (5)$$

where n_i denotes the number of particles of volume i, and n_j, the number of particles of volume j.

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Unfortunately, approximately 2 × 106 ordinary differential equations would be required to describe the anticipated range of particle volumes in this system. To simplify the numerical implementation of the solution, a geometric sectionalization method was used with a geometric ratio of 2.[2, 3] Here, the geometric ratio is defined as

$$\frac{v_{i+1}}{v_i} = 2^{1/q} \tag{6}$$

where v_i is the lower bound of the ith volume interval, v_{i+1} is the lower bound of the next greater volume interval, and q is the discretization parameter, taken in this case to be 1. This method works well with the binary equisized daughter distribution assumed for particle breakage, and the method has been shown to capture particle number and particle mass balances correctly. It should be noted that for values of q, greater than 4, corrections to the original population balance discretization approach of Litster, *et al.*,[2] were published by Wynn.[3]

The number of bins was left to the students, but it was suggested that the students cap the number at 28 bins. Detailed comparisons between predictions of size distribution evolution made using this sectional approach with 25 bins and predictions made using the quadrature method of moments and the polynomial interpolative closure method of moments have been reported previously.[4]

The population balance equations used to describe the change in the number of particles in bin i (N_i) as a function of distance down the axis in the sectional approach are given by:

$$u_z \frac{dN_i}{dZ} = N_{i-1} \sum_{j=1}^{i-2} \frac{\beta_{i-1,j}}{2^{i-j-1}} N_j + \frac{1}{2} \beta_{i-1,i-1} N_{i-1}^2 - N_i \left[\sum_{j=1}^{i-1} \frac{\beta_{i,j}}{2^{i-j}} N_j + \sum_{j=1}^{\infty} \beta_{i,j} N_j \right] + 2\Gamma_{i+1} N_{i+1} - \Gamma_i N_i \tag{7}$$

with the collision kernel given by:

$$\beta(i,j) = \frac{2kT}{3\mu}\left[2 + 2^{(i-j)/3} + 2^{(j-i)/3}\right] + 0.31\sqrt{\frac{\varepsilon}{\nu}}\left(\frac{3V_0}{2}\right)\left[2^{i-1} + 3\left(2^{(2i+j)/3-1} + 2^{(i+2j)/3-1}\right) + 2^{j-1}\right] \tag{8}$$

To convert to particle diameter from the volume distribution—since the particles are colliding at low temperatures to form soft agglomerates—it was assumed that the agglomerates were fractal-like, and therefore particle volume scaled with particle diameter, d_p, as:

$$d_p = d_o \left(\frac{6V}{\pi d_o^3} \right)^{\frac{1}{D_f}} \tag{9}$$

where d_o is the primary particle diameter, and D_f is the fractal dimension, here assumed to be 1.8.

PROBLEM IMPLEMENTATION

The design assignment followed lectures on particle size distributions and fractal aggregates, diffusion, nucleation, coagulation, coalescence, discrete population balance modeling approaches, and cyclone design by the course instructor (Ehrman). The industrial partner (Diemer) gave a lecture on industrial applications of aerosols, and on sectional population balance approaches specific to the design problem.

First Implementation, Spring 2003

In the first problem assignment, in 2003, students were asked only to design the coagulation tube. The cyclone grade efficiency, η, was given as:

$$\eta = \frac{\left(d_p / d_{pc}\right)^2}{1 + \left(d_p / d_{pc}\right)^2} \tag{10}$$

where the critical particle diameter, d_{pc}, or cut size, was 24 microns. The maximum pressure drop allowed in the coagulation tube, 2 psi, was also given as a constraint. The students were restricted

to integral values, in inches, for the pipe inner diameter. The students were asked to write a program to solve the population balance equations, using a program of their choice. Global optimization of this problem was beyond the scope of the course and so solution approaches involved trial and error selection of pipeline agglomerator diameter and length, and from the final size distribution, the cyclone mass capture percentage was calculated from Eq. (10). This process was repeated until a solution meeting all specifications and satisfying all constraints was identified.

Students worked on this problem in teams consisting of three or four students. The students were given three weeks to complete the problem. Students were given a chance to ask the industrial partner questions in a conference call one week before the project was due, and two teams participated in this call. One team was successful at writing a code in MATLAB to solve the population balance equations, and they used a systematic approach to find pipeline agglomerator dimensions that allowed for adequate agglomeration, yet minimized pressure drop. The solution format was a brief three-page industrial-type memo, addressed to the instructor and industrial partner, and describing the final design and the particle size distributions entering and exiting the cyclone. Two teams were not able to finish their code in the time allotted. These teams were asked to describe the solution algorithm they would have followed if they had gotten their code to work. Students were given the opportunity to submit evaluations of the class at the midterm point anonymously through our Web-based course-hosting software, and the following comment is representative of many of the student's views about the project.

> *"I think the computer project was a bit over our heads. We could program the algorithm as outlined by Dr. Diemer, but it was more or less an exercise in MATLAB instead of population balance modeling."*
>
> *— anonymous student, spring semester, 2003*

As a result of the project outcome, the project was modified significantly before its second implementation.

Second Implementation, Fall 2005

The main difference with the second implementation is that students were given a Matlab code to solve the population balance equations, written by the group of students who successfully completed the problem in 2003. To increase the degrees of freedom in the problem, the design portion was expanded to include the cyclone. Students were asked to design the cyclone using ChemCad software, rather than being given a grade-efficiency curve and cut size for a specific cyclone. With the incorporation of the process simulator, the intent was to add a secondary aspect to the project: giving students an experience extending chemical engineering process simulation software to particle technology. Students were asked to minimize the area of the pipeline agglomerator and cyclone, as a surrogate for minimizing capital costs. The students were given a constraint of a maximum pressure drop in both pipeline agglomerator and cyclone of 2 psi. The preparation for this implementation was similar to the first one, with one exception. Prior to Dr. Diemer's lecture, the teaching assistant (Dwivedi) gave a hands-on tutorial on cyclone design using ChemCad, in our computer classroom. Teams were assigned such that at least one person on a team had prior ChemCad experience. Prior to this project, however, these students had not previously worked with multiphase process streams consisting of suspended particles, so a tutorial for all students was a necessity.

An additional individual homework assignment was given prior to the project being assigned: Students were required to calculate the magnitude of each component of the coagulation kernels over a range of particle diameters. The goal was to get a feel for the relative importance of continuum Brownian vs. turbulent coagulation, and how the relative importance of each changed as particle size increased. Additionally, this assignment gave some assurance that all students were comfortable with the collision kernel calculations.

In the design problem itself, the basic solution algorithm most groups followed was:

1. *Choose a length and diameter for the pipeline agglomerator.*
2. *Run the MATLAB code to determine the size distribution at the end of the agglomerator.*
3. *Input the size distribution, as well as the cyclone size specifications into ChemCad and calculate the composition and size distribution of the cyclone exit stream.*
4. *Calculate the overall pressure drop in the system and check to see if the constraint of 2 psi is satisfied; if not, return to step 1 or step 3.*
5. *Calculate the surface area of the agglomerator and cyclone.*
6. *If pressure drop was significantly below 2 psi, reduce cyclone or agglomerator dimensions and return to step 2.*

There was one exception to the basic algorithm described above. One group focused on the cyclone and designed an extremely small cyclone, with a pressure drop of nearly 2 psi that had high efficiency for micron-sized particles, and thus they only required a very short pipeline agglomerator. Five out of six groups were able to complete the project in two weeks. The teaching assistant held additional office hours to assist with the process simulator and the instructor held office hours to assist students in interpreting and modifying the MATLAB code. The students did not request a conference call with the industrial partner. As in 2003, the solution was submitted in the form of a brief three-page industrial memo containing the details of the design, the size distribution of particles entering

the cyclone in histogram format, and the size distribution and solids mass fraction of gas leaving the cyclone.

Anonymous feedback in the second implementation was generally more positive than in 2003. The students "enjoyed learning about some of the related new developments in the industry concerning major companies like DuPont," and they enjoyed working on a realistic, challenging, open-ended problem. There was considerable relief that the MATLAB code for the sectional solution to the population balance equations was supplied. There was, however, one comment suggesting that it would be better to "let students make [the] population balance model by themselves."

CONCLUSIONS

With the modifications made in the second implementation, we feel we have created an interesting and tractable design problem. We intend to continue including this assignment, with slight modifications, as an integral part of this course in the future. Detailed 2003 and 2005 problem statements as well as the MATLAB code, and a guide to ChemCad and cyclones, are available at: <http://www.glue.umd.edu/~sehrman/popbal.htm>.

ACKNOWLEDGMENTS

S.H. Ehrman acknowledges the support of a CAREER award from the National Science Foundation (DMR 0093649) supporting educational advances in the field of particle technology and materials processing.

REFERENCES

1. Nelson, R.D., R. Davies, and K. Jacob, "Teach -em Particle Technology," *Chem. Eng. Ed.*, , **29**(1), 12 (1995)
2. Litster, J.D., D.J. Smit, and M.J. Hounslow, "Adjustable Discretized Population Balance for Growth and Aggregation, "*AIChE Journal*, **41**(3), 591 (1995)
3. Wynn, E.J.W., "Improved Accuracy and Convergence of the Discretized Population Balance of Litster, *et al.*," *AIChE Journal*, **42**(7), 2084 (1996)
4. Diemer, R.B., and S.H. Ehrman, "Pipeline Agglomerator Design as a Model Test Case," *Powder Technology*, **156**, 129-145 (2005) ❐