

A course in . . .

PARTICULATE PROCESSES

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A USEFUL WORKING definition for particles [1] is, "that state of subdivision of matter whose shape depends on the process by which it was formed and on the intermolecular cohesive forces present." This definition applies equally well for liquid droplets (spherical, maintained by surface tension) or crystalline solids having a geometric shape (e.g., cube, platelet, etc.) consistent with the crystalline structure and affected by the local molecular environment producing the crystal.

This article describes a special topics graduate course (ChE-514) on particulate processes given frequently by the author at the University of Arizona. The text for the course is *Theory of Particulate Processes: Analysis and Techniques of Continuous Crystallization* [2]. The subtitle has been said to be more accurate in describing the book than the title, although the second edition attempts to correct this impression. The text was motivated by the necessity of collecting and organizing all the information on the Crystal Size Distribution (CSD) problem, which is covered extensively in the course Particulate Processes. Thus, the course and text are nearly inseparable. ChE-514 is a "required" course for the writer's students who are engaged in process crystallization research. The course is given whenever the combination of graduate students needing to take it (ADR's) plus other graduate students desiring additional chemical engineering credit to fill out their graduate study pro-

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TABLE 1
Course Topics for Particulate Processes

- Introduction and Motivation: The Importance of PSD/CSD
- Particle Distributions
- The Population Balance
- Modeling Continuous and Batch Crystallizers
- Crystallization Kinetics
- Crystal Size Responses for Continuous and Batch Crystallizers
- Reaction Engineering of CSD
- CSD Dynamics and Control

gram, exceeds the minimum class enrollment for a graduate offering. The course unashamedly concentrates on process crystallization (and CSD) as the example *par excellence* to illustrate the predictive population balance theory of particulate processes formally developed in the text. The writer attempts to maintain a reasonable balance of non-crystallization topics considering the background of those enrolled.

COURSE OUTLINE

Table 1 shows the subject outline of Particulate Processes. It is identical to the text with the exception of Chapter 10 (in the course, the last periods are used for student reviews of the current literature of particulates). The ground rules are that crystallization students cannot choose a crystallization article to review, while others may. The main point is that the articles must emphasize the distributed nature of particulate systems. Proposed titles are thus pre-screened. Five minutes of perusing the article to be reviewed can immediately determine if the course has been a success.

One graduate student suggested that scarce semester-end time could be saved if written, rather than oral, critiques were handed in as a term project. This is an excellent idea except, of course, that it shifts a major work load from the student to the in-

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structor. In addition to emphasizing the distributed nature of particulates, the course emphasizes predictive rather than descriptive modeling of the particle distribution using population balance mechanics. An illustration of CSD prediction and manipulation will be presented for the useful double draw-off (DDO) configuration. Current unit operation texts [3] now present the CSD from a Mixed Suspension, Mixed Product Removal (MSMPR) crystallizer in a predictive context, but stop short of CSD manipulation (which would require crystallization kinetics). The first two chapters (and course topics) describe the general nature and elementary statistics of distributed particulates (*e.g.*, means, variance, cumulative

vs. density, etc.). The distributions are presented in density form. (Students often have trouble with the units of population density, $(\text{length})^{-4}$). Much attention is given to the gamma distribution (the natural distribution of crystallization processes), but other useful empirical distributions, *e.g.*, Rosin-Rammler and Gaudin-Melloy, that are routinely used in the minerals industry [4] are presented in the course.

Chapter 3 develops and formalizes the multi-variate population balance which is used predictively throughout the remainder of the course. At this point, the useful moment transformation is introduced. The leading moments of the population density function

$$\left[m_j = \int_0^{\infty} L^j n(L) dL \right] \text{ for } j = 0, 1, 2, 3$$

form a closed set of non-linear algebraic equations which, in principle, completely describe the idealized MSMPR crystallizer, given the nucleation/growth rate kinetics of a particular system. Roughly speaking, the MSMPR concept is to crystallization as the CSTR is to reaction engineering with the advantage that the *form* of the equations is kinetics-independent. Thus, for a specific case the kinetics can be brought in as auxiliary equations to complete the solution.

Chapter 4 develops the MSMPR concept in detail. This chapter, together with Chapters 7 (CSD manipulation) and 8 (CSD dynamics), form the core of presentations for the industrial short course. Chapter 5 presents crystallization mechanisms and kinetics from an elementary level. The writer often suffers from acute Felder's Impostor Syndrome [5] when discussing crystal nucleation and growth mechanisms. This subject could better be covered by someone in material sciences well-versed in crystallography. For example, when discussing crystal growth mechanisms by spiral dislocations, the writer finds that even the most imaginative students are barely convinced that the crystal dislocation is self-perpetuating. Crystal nucleation/growth kinetics can often be described for high yield systems with simple power-law empiricisms of the form

$$B^{\circ} = k_N G^i M_T^j$$

where *i* and *j* are two parameters respectively describing
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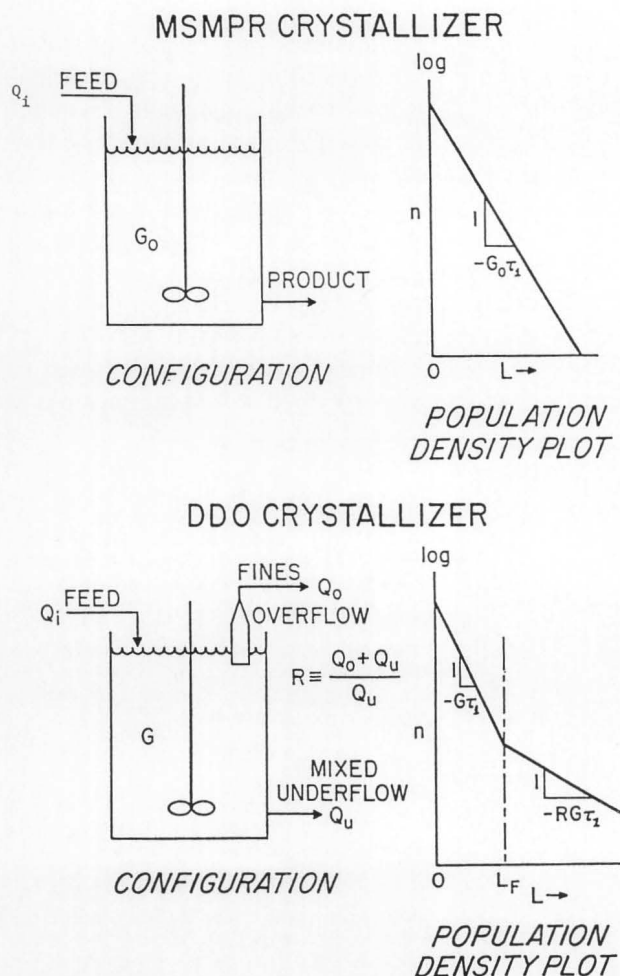


FIGURE 1. MSMPR and DDO configurations and CSD (after E. T. White and A. D. Randolph (1988)).

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ing the relative sensitivity of secondary nucleation to growth rate G (used as a surrogate variable for supersaturation) and slurry density M_T .

CSD SIMULATION AND MANIPULATION

Figure 1 shows the configuration and theoretical population density plot for both the MSMPR and Double Draw-Off (DDO) crystallizers [6]. The DDO configuration merely removes and then combines two separate slurry streams, one mixed and one classified to

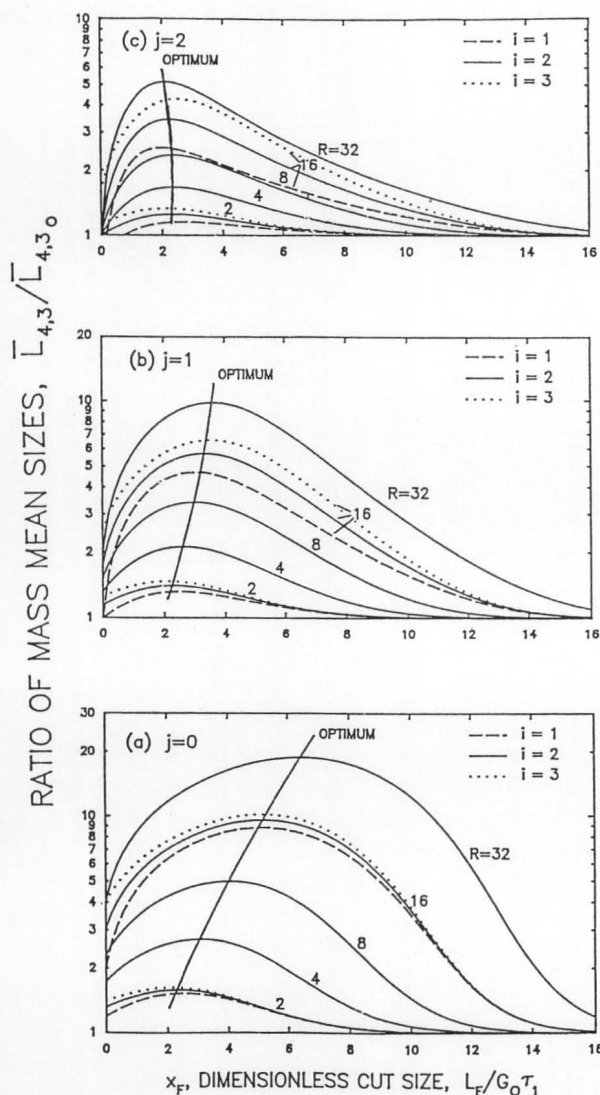


FIGURE 2. Mass Mean Size Improvement, DDO/MSMPR Crystallizers (after E. T. White and A. D. Randolph (1988)).

contain only crystals less than some cut size L_F . Classification is usually done passively by settling within the vessel. Figure 2 shows the dramatic average particle size increase that this simple configuration can achieve *vis-a-vis* the MSMPR configuration. Simple power-law nucleation kinetics of the form

$$B^{\circ} = k_N G^i M_T^j$$

were used for these calculations. As the slurry density also increases in DDO operation this configuration is only fully useful for weak feeds giving a low natural slurry density. Per-pass yield is also increased. Thus, the DDO configuration is also used to increase yield in systems with slow growth kinetics.

Bench-scale studies are currently being done to evaluate the DDO crystallizer as a method of making larger calcium sulfite and sulfate (gypsum) particles in Flue Gas Desulfurization (FGD) processes. Larger particles would greatly reduce downstream costs in such FGD processes.

In ChE-514, students have access to a computer program (Program Crystal Ball [7]) which solves simultaneous population and mass balances for the CSD using arbitrary crystallization kinetics. Students use this program to design a crystallizer producing a desired crystal size and yield.

In summary, the course explores the PSD of particulate processes, while emphasizing the distributed nature of these processes. It attempts to show prediction as well as description of the PSD with the ultimate aim of manipulation. However, these goals are only achieved in the study of CSD from well-defined crystallization processes.

REFERENCES

1. Irani, R.R., and C.F. Callis, *Particle Size: Measurement, Interpretation, and Application*, John Wiley & Sons, New York (1963)
2. Randolph, A.D., and M.A. Larson, *Theory of Particulate Processes: Analysis and Techniques of Continuous Crystallization*, second edition, Academic Press, San Diego, CA (1988)
3. McCabe, W.L., J.C. Smith, and P. Harriott, *Unit Operations of Chemical Engineering*, fourth edition, McGraw-Hill, New York (1985)
4. Kelly, E.G., and D.J. Spottiswood, *Introduction to Mineral Processing*, Wiley, New York (1982)
5. Felder, R.M., "Impostors Everywhere," *Chem. Eng. Ed.*, **22**, 168 (1988)
6. White, E.T., and A.D. Randolph, "Optimum Fines Size for Classification in Double Draw-Off Crystallizers," *Ind. Eng. Chem. Res.*, **28**, 276 (1989)
7. Sharnez, Riswan, "Dynamic Simulation and Control of Crystal-Size Distribution in a Continuous Crystallizer," MS Thesis, University of Arizona (1987) □