

A Graduate Course In

Chemical Reactor Engineering*

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We need hardly consider the necessity for graduate instruction in Chemical Reactor Engineering; various surveys and intuition itself suggests that training in this area is vitally necessary to the practicing chemical engineer. Further, given certain ecumenical trends in other disciplines, e.g., thermodynamics, heat transfer, etc., the uniqueness of chemical engineering may well be preserved only to the extent that chemical reactor engineering becomes the central focal point of academic concern.

Table I lists the recommended texts and Table II gives the topics embraced by the course I teach to first year graduate students at Notre Dame. Two things are evident, (1) little emphasis is

TABLE I.

Texts Recommended for Course in Chemical Reactor Engineering

1. Kramers, H., and K. P. Westerterp, "Elements of Chemical Reactor Design and Operation," Academic Press, New York (1963). It has broad coverage, but lacks problems and the illustrative examples lack detail.
2. Denbigh, K., "Chemical Reactor Theory," Cambridge University Press, New York (1965). An excellent book, but it is best used after students gain some familiarity with more detailed texts.
3. Levenspiel, O., "Chemical Reaction Engineering," Wiley, New York, (1962). This text is very good for those who come to us with a weak background in chemical engineering kinetics.
4. Smith, J. M., "Chemical Engineering Kinetics," McGraw-Hill, New York, (1956). This text has excellent problems and well detailed examples.
5. Petersen, E. E., "Chemical Reaction Analysis," Prentice-Hall, New York (1965). This text is very good on heterogeneous reactor problems.

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TABLE II.

Course Outline for Chemical Reactor Engineering

1. Chemical kinetics review and data treatment
Conversion and yield in simple, complex, and autocatalytic reactions. 3 periods
2. Reactor types—CSTR, PFR, and recycle RXR
Isothermal and adiabatic behavior in relation to conversion and yield. Reference: *Chem. Eng. Sci.* 21, 472 (1966); *I&EC (Fund)* 5, 164 (1966). 4 periods
3. Continuity equations and parameter correlations
Reduction of continuity equations to specific reactor types. Theory and data for key physical parameters. Boundary conditions, simplifications, and scale up. Reference: *I&EC (Nov.)* (1964); *Can. J. Chem. Eng.* 36, 207 (1958). 5 periods
4. Homogeneous reactor design
Iso- and non-isothermal cases. Thermal stability and transient analysis (n-CSTR in series). 5 periods
5. Heterogeneous systems (gas-liquid reactions)
Film vs. penetration model. The van Krevelen plot and its use by Andrew. Reference: *Brit. Chem. Eng. (Jan./Feb.)* (1967); *Chem. Eng. Sci.* 21, 1951 (1966). 3 periods
6. Gas-solid non-catalytic reaction
Shell progressive mechanism. Reference: *Can. J. Chem. Eng.* 43, 334 (1965). 2 periods
7. Heterogeneous catalysis
Physical and chemisorption; rate and equilibrium hetero/homogeneous surfaces and catalytic rate models derived therefrom. Data analysis and ambiguity of models. Laboratory reactors. 10 periods
8. Catalytic reactor design and analysis of fixed and fluidized beds. Inter/intraphase diffusion of heat and mass. Influence of this diffusion on conversion and yield. Reactor simulation of non-isothermal and adiabatic fixed beds—SO₂ oxidation on Pt and V₂O₅; styrene production, organic oxidation. Two phase model of fluidization. Reference: *I&EC (Oct.)* (1966); *AIChE J.* 6, 460 (1960) and 9, 129 (1963); *Chem. Eng. Sci.* 17, 675 (1962). 13 periods

placed on chemical kinetics *per se* and, (2) major emphasis is given to heterogeneous systems. Concerning point (1), our belief is that chemical kinetics is best taught in our Chemistry Department. On point (2), I endorse the wise observation of Olaf Hougen, to wit: the most commonly encountered and challenging reactor problems are those of a heterogeneous nature. Thus, perhaps 75% of our course is devoted to heterogeneous reaction-reactor problems.

Following a brief review of chemical kinetics and data treatment wherein emphasis is placed on yield/selectivity, we consider the reactor problem. This, I feel, is best treated by developing the continuity equations for a non-isothermal/non-adiabatic reactor as shown in Table 3. These equations must be reducible to various reactor types and environments, e.g., CSTR, PFR semi-batch, etc. Logically, at this point, the phenomenological coefficients of axial and radial dispersion (heat and mass) are discussed and the various correlations set forth with supporting a *a priori* theory for both packed and unpacked tubes.

Thus, the mixing and residence time issue is introduced in terms of the axial Peclet number. I devote little time to residence time distribution problems as I contend that non-isothermality in both axial and radial directions proves far more telling upon conversion and yield than does back-mixing and/or by-passing.

Table III.

Steady State Continuity Equations

Mass :

$$\frac{\partial f}{\partial n} - \frac{1}{d_p u/D_r} \left[\frac{\partial^2 f}{\partial m^2} + \frac{1}{m} \frac{\partial f}{\partial m} \right] - \frac{1}{d_p u/D_a} \left[\frac{\partial^2 f}{\partial n^2} \right] = \frac{\theta_a \Sigma R}{C_o n}$$

(a) (b) (c) (d)

Temperature :

$$\frac{\partial t}{\partial n} - \frac{1}{d_p u/k_r} \left[\frac{\partial^2 t}{\partial m^2} + \frac{1}{m} \frac{\partial t}{\partial m} \right] - \frac{1}{d_p u/k_a} \left[\frac{\partial^2 t}{\partial n^2} \right] = \frac{\theta_a \Sigma (-\Delta H) R}{\rho C_p T_o n}$$

where $f = C/C_o$; $t = T/T_o$; $n = z/L$; $m = r/R_o$

PFR : $a=d$; CSTR : $a(A)=d$;
PFR with dispersion : $a+c=d$; Non-iso PFR : $a+b=d$.

In general

Mass :

$$\frac{1}{V} \frac{\partial cV}{\partial \theta} + u \frac{\partial c}{\partial z} - D_r \left[\frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial c}{\partial r} \right) \right] \dots = R = rxn + add'n$$

readily reduces to PFR; batch and semi-batch (variable volume).

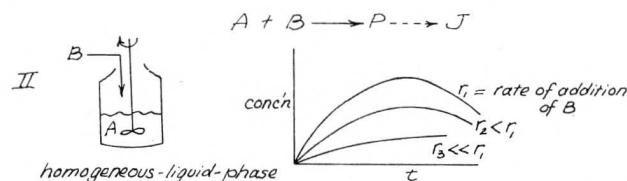
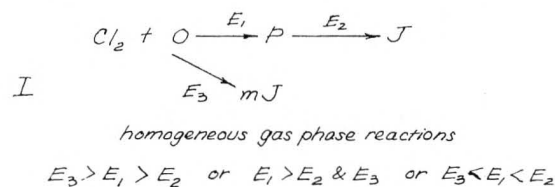
HOMOGENEOUS SYSTEMS

We next consider homogeneous design problems (see Table 4). I rather like this consecutive simultaneous network. Each student is assigned such a problem so that by numerical computation, the class witnesses precisely how yield is affected, under non-isothermal/non-adiabatic conditions, by physical parameters (flow rate of process and coolant streams, feed composition, etc.) as well as chemical parameters (activation energies and their relative magnitudes).

Semi-batch design problems are also assigned. Save for illustrating mixing models, I devote little time to CSTR networks. Dynamic behaviour is readily explored at this point by reviewing the n-CSTR series treatment under unsteady state conditions.

Table IV.

Typical Homogeneous Reactor Design Problems



III Phosgene decomposition - homogeneous
Adiabatic - use Douglas - Eagleton Exp. Integral

HETEROGENEOUS SYSTEMS

I treat the gas-liquid reaction problem first. This has the advantage of avoiding detailed mechanistic inquiry (required of heterogeneous catalysis). Table 5 indicates two types of gas-liquid reaction problems. The first, CO₂ absorption in NH₃ solution, illustrates the power of the van Krevelan plot and Andrew's analysis of it. *A priori* design is possible here. In the second case, design is based on use of a semi-empirical

Table V.

Gas-Liquid Reactions

I CO_2 (0.1 atm) + $10^{-3} \frac{\text{gmole}}{\text{cc}}$ NH_3 solution on a sieve tray. Determine plate efficiency

k_L determined in laboratory where α' known $\Rightarrow R_r$
 α' determined for sieve tray (CO_2 absorbed into NaOH)

von Krevelen plot yields k_L/k_{L0} for sieve

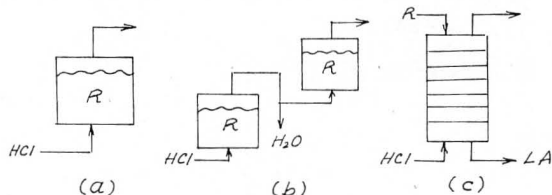
Reference: Andrew, CES, 3, 279 (1954)

Bridgwater

Brit. Chem. Eng., Jan & Feb., 1967

II ROH (Liquid) + HCl (Gas) \rightarrow Lauryl Chloride

Problem: Find conversion and HCl utilization for



Kingsley & Bliss (I&EC, 44, 2479 (1952)); $r = k \left(\frac{G}{V}\right)^{0.5} \sqrt{\text{ROH}}$

laboratory secured rate law. Details of each are presented in the January and February issues of *British Chemical Engineering* (1967). Gas-solid non-catalytic reaction is next treated. The shell-progressive mechanism is involved after its validity is justified. The isothermal case for changing particle size is handled as described by White in *Can. J. of Chem. Eng.* 43, 334, (1965). Non-isothermal analysis follows the treatment of Smith and Aris's generalization.

The final phase of the course is concerned with catalysis *per se* and catalytic reactor design. I devote several lectures to physical-chemical adsorption rates and equilibria, emphasizing the ideal-non-ideal surface implications with particular reference to rate models derived therefrom. We conclude that the word "mechanism" never be used: "model" is to be preferred. Exercises in data treatment are assigned. I refer the students to the literature on non-linear least square analyses.

Finally both fixed and fluid bed design problems are faced. Diffusional aspects (heat and mass) both about and within the catalyst are treated and we then discuss computer simulation. Figure 1 reveals very nicely how both inter-intra-phase diffusion phenomena affect intrinsic chemical dispositions in the adiabatic catalytic oxidation of SO_2 .

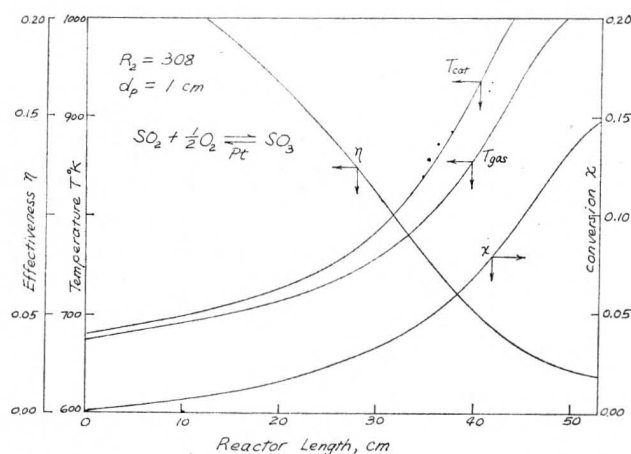


Fig. 1.—Diffusion phenomena affect intrinsic chemical dispositions.

Unfortunately I find no time to cover such topics as ion exchange, GLC and GSC. The important area of optimization is treated more effectively in a companion course taught by my colleague Dr. Crandall.

"I go to seek a great perhaps . . .," Rabelais remarked. In the design of heterogeneous reactors, we'd do well to keep this comment in mind—particularly in the minds of our students.



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