Changing Attitudes

TO REACTOR DESIGN

Octave Levenspiel

Professor of Chemical Engineering
Illinois Institute of Technology, Chicago, Illinois

Exciting things are happening today in the field loosely covered by the terms *chemical technology*, *reactor design*, and *kinetics*, for, without fanfare, study reports, or discussion in ASEE or in AIChE, a new way of looking at this subject was recently introduced. The new view may be characterized by the phrase *Chemical Reaction Engineering*, and its friendly reception by the profession is causing a remarkable change in our approach to and our teaching of chemical technology.

I should like to discuss this new development, to sketch its strategy as opposed to the traditional approach, to give a few examples to illustrate what I mean, to show how it has affected our pattern of education, and finally to consider what I feel is its place in the main line evolution of our profession.

In the traditional approach, we engineers were more concerned with the peculiar and unique features of this or that reacting system than with the similarities and common characteristics among such systems. It is true that the unique features make for the success of a particular process. It is also true that what is useful for the processes of tomorrow are the analogies and generalizations which we are able to extract from the technology of today. Thus for predictive and design purposes we need generalizations; in the past, however, we have emphasized the particular and the unique.

In the U.S. of the forties we saw the first break with this philosophy. The general approach was brought forward; however, it was soon channelled, narrowed, and dominated by the problems and interests of the oil industry. Hence there was an overwhelming emphasis on catalytic reactions and a rather unhealthy preoccupation with developing models to account for their kinetics. Little wonder that this subject was known and is still known as *applied kinetics*, *chemical engineering kinetics*, or just *kinetics*, despite the fact that finding

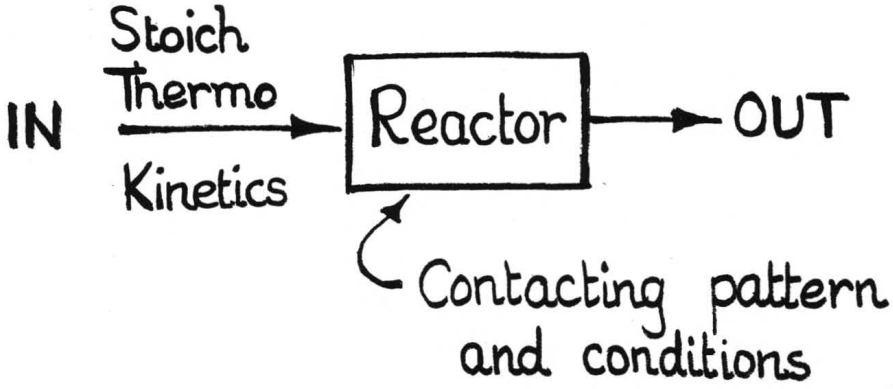


Fig. 1 — Schematic of Chemical Reactor Engineering

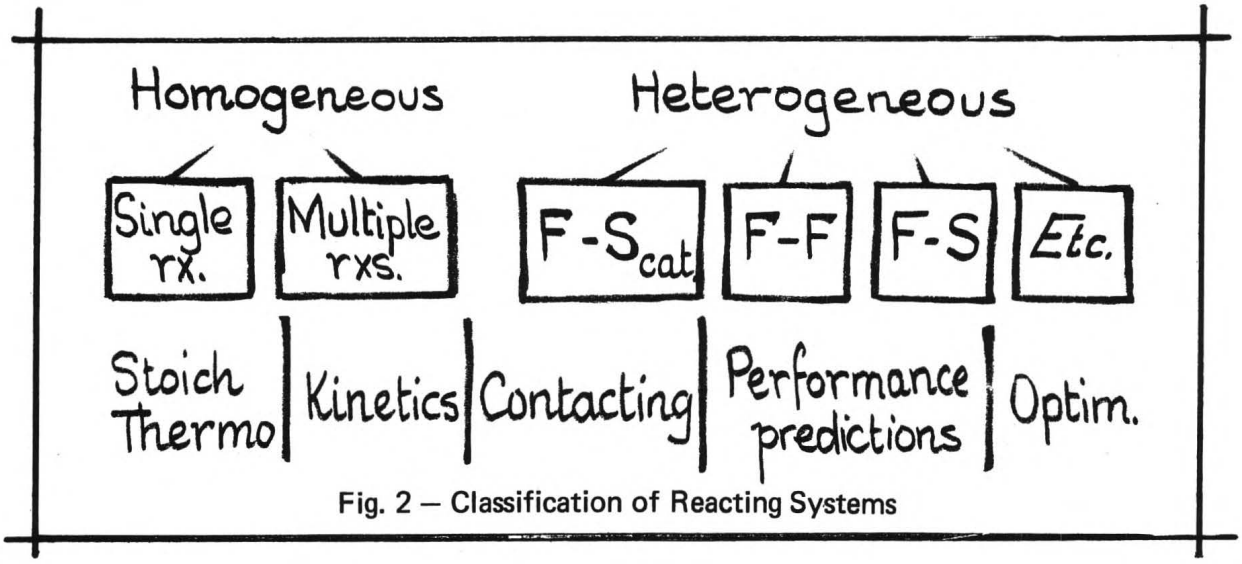


Fig. 2 — Classification of Reacting Systems

the rate of reaction is only one phase in the over-all problem of scale-up and proper reactor design.

In the late fifties a bolder attempt at generalization was started in Europe. To emphasize its concern with the over-all problem it was termed *Chemical Reaction Engineering* or CRE for short. As the name suggests, we are here concerned with the "engineering" of chemical reactions, or their efficient exploitation. The philosophy, then, is to seek common factors to link processes and to develop broad methods of attack for problems, and this is done with the conviction that these methods can be used as a basis for design in *all* areas of application, whether it be in polymer processing, industrial metallurgy, pharmaceutical manufacture, foods, biological processes, high-temperature reactions, or any other operation — in fact wherever a new material or chemical species is to be produced economically on a commercial scale.

In a nutshell the concern of CRE is shown in Figure 1. Thus we engineers must determine how to treat a feed stream so as to obtain most economically a most desirable product stream. More specifically we must decide what temperature, pressure, and contacting pattern to use, and in general this decision requires knowledge of the stoichiometry, thermodynamics and kinetics of the systems.

It is convenient to classify reacting systems according to the phases participating in the reaction. As shown in Figure 2 the broad division is between homogeneous and heterogeneous systems. In the former, one fluid phase alone moves through the reactor and only chemical kinetic factors influence the rate. In the latter classification, we may have to deal with additional problems arising as a direct consequence of the heterogeneity of the system, such as phase equilibrium, mass transfer from phase to phase or from phase to interface, and contacting pattern of the phases. As may be expected, heterogeneous systems represent a more complex situation.

Next we see that homogeneous systems may be divided into systems where only one reaction occurs and into systems where two or more reactions occur simultaneously. The reason for this split is that different questions are pertinent in these two situations. With one reaction all we ask is to maximize conversion with a most economical reactor arrangement; with multiple reactions we ask how (by what temperature, pressure, and contacting pattern) to promote the formation of the desired product and depress the formation of all other undesired materials. The primary concern is not the same in the two cases. And so it is with heterogeneous systems, where we have (in what I consider to be the order of complexity) solid-catalyzed gas-phase reactions, non-catalyzed fluid-solid reactions (reduction of metal oxides, burning of coal), fluid-fluid systems (absorption with reaction), and finally the catch-all *et cetera* classification which includes all other multiphase systems. These systems can be horribly messy to treat, and in the interest of simplicity I shall avoid them in this paper, even though they include many processes of great industrial importance.

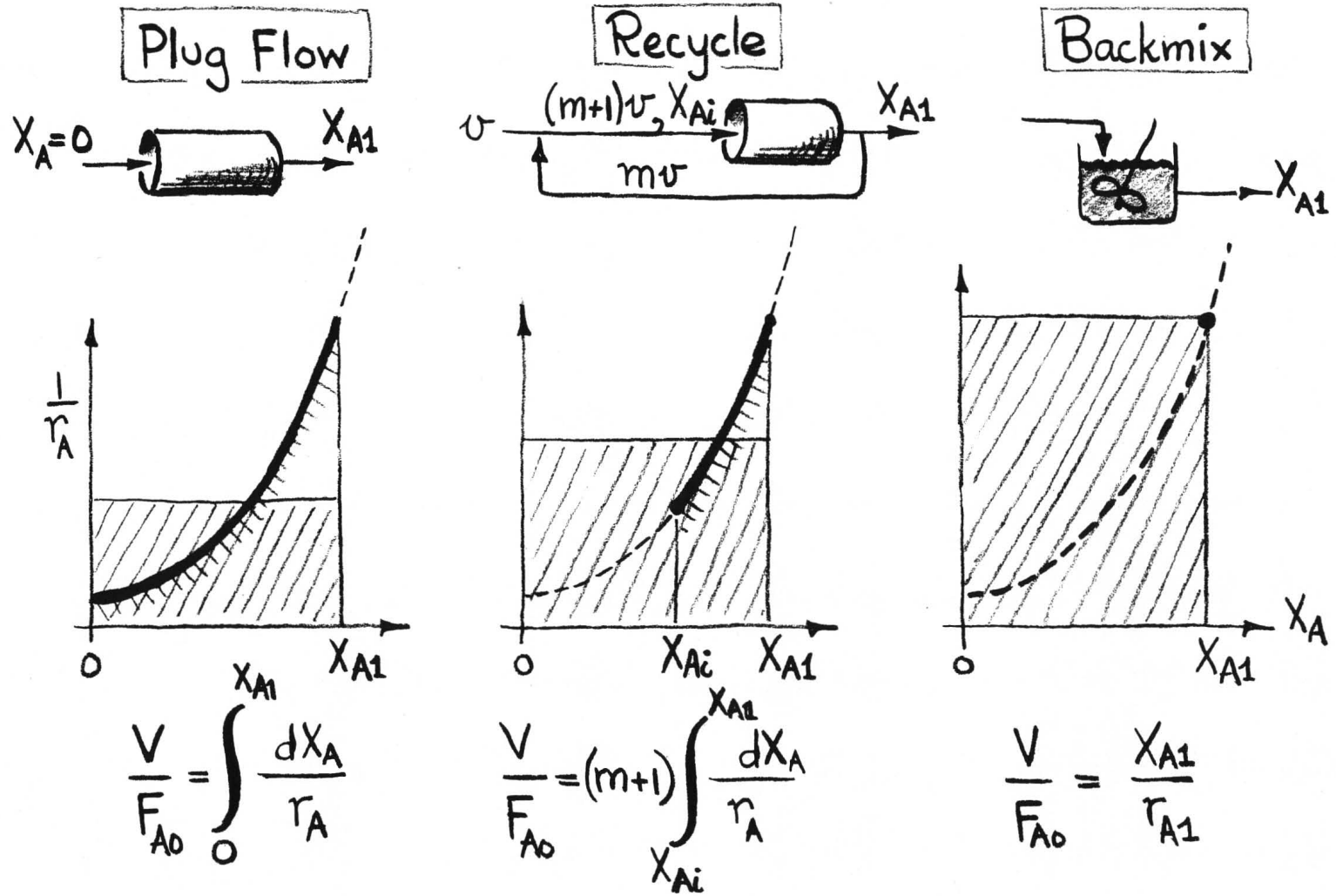


Fig. 3 — Reactor Flow Patterns: Design Equations

The line at the bottom of Figure 2 shows the usual stages in our control of reacting systems all the way from initial studies on thermodynamics and stoichiometry to a search for optimum operation.

Our ability to handle problems is quite different in the different classifications. For example in homogeneous single-reaction systems we are at the stage of selecting optimum set-ups. In catalytic systems we would be just about as far along if it were not for the obstinate problem of representing and properly accounting for the kinetics of the reaction. With fluid-fluid systems our difficulties are everywhere: how to represent the rate usefully, how to account for movement of reacting species in the flowing phases, and in general how to predict the extent of reaction given all kinetic and equilibrium information. This means that except for special cases we have trouble trying to develop a design equation for this classification. And so it is with the other classifications shown in Figure 2; in each there are particular difficulties to be overcome.

Let me next offer three examples of recent research activities in CRE. They must be brief for this review to conform to reasonable length. Nevertheless I hope that they will provide a feeling for the sort of thing that is going on.

The first example concerns the search for an optimum set-up for the single catalylic reaction whose thermodynamics and stoichiometry are known, and let us restrict ourselves to adiabatic operations in packed beds. Now there are many possible ways of running such a reaction — for example, single-stage operations in one packed bed reactor, multistage packed-bed operations with interstage heat transfer, operations with recycle of fluid between stages or with bypassing of certain stages with fresh feed. And for every set-up we can choose the temperature of fluid entering the various stages. With all these factors at our control, are we to make a computer search of every possibility that comes to mind, or are there methods which give us a feel for the situation and allow us to reject quickly many of the alternatives while retaining those few which are promising? Since I favor the latter approach let me outline a graphical method which does just that.

Figure 3 shows the design equations for the flow patterns of interest: the ideals of plug flow and backmix flow and the inter-

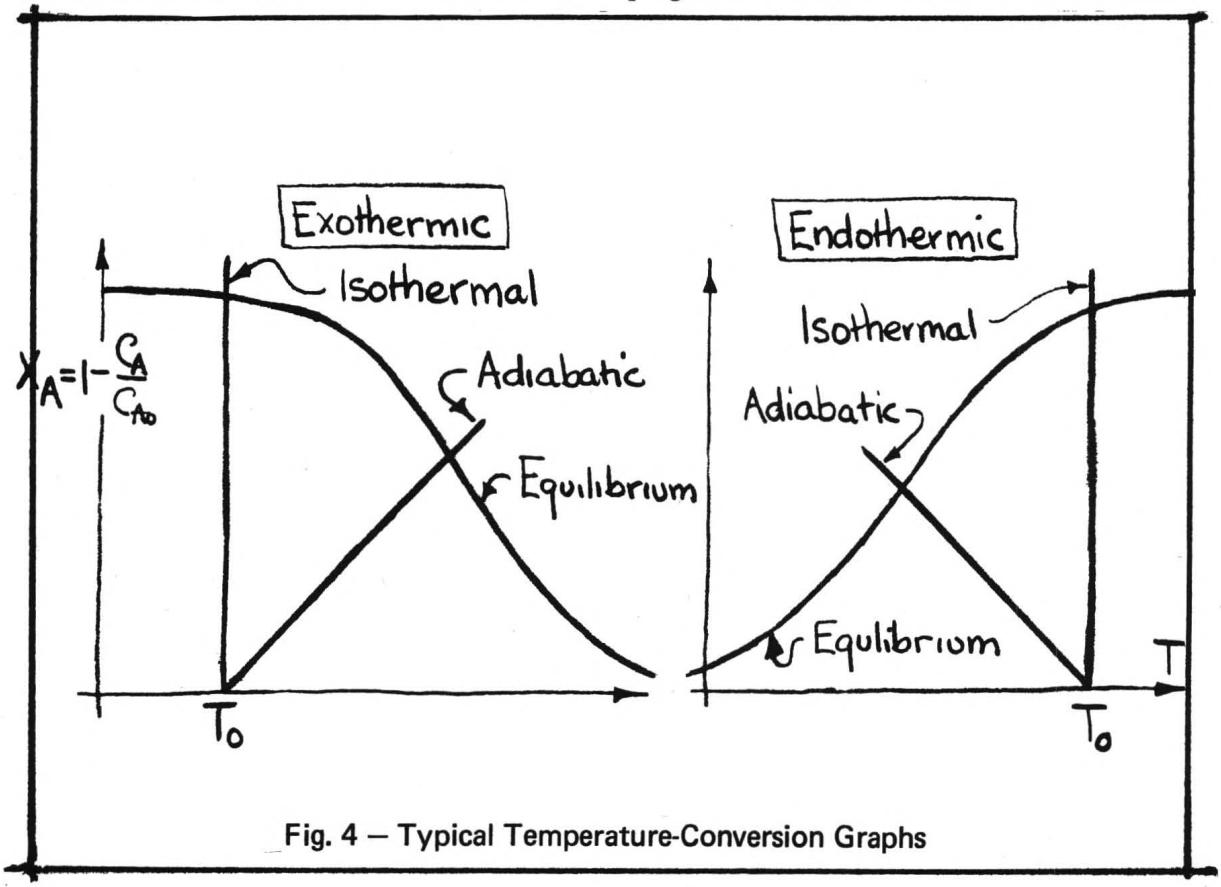


Fig. 4 — Typical Temperature-Conversion Graphs

mediate of recycle flow. It should be noted that plug and backmix flows represent the extremes of recycle flow when the recycle ratio m becomes zero and infinite, respectively. These equations indicate that the four quantities, V (size of reactor), F_{A0} (feed rate of reactant A), $-r_A$ (rate of reaction), and X_A (conversion of reaction) are all interrelated, and knowing any three of these quantities gives the fourth. The sketches in Figure 3 are a graphical representation of these equations for any arbitary kinetics and the shaded areas are a measure of the size of reactor needed for a given duty. These expressions are quite basic to CRE.

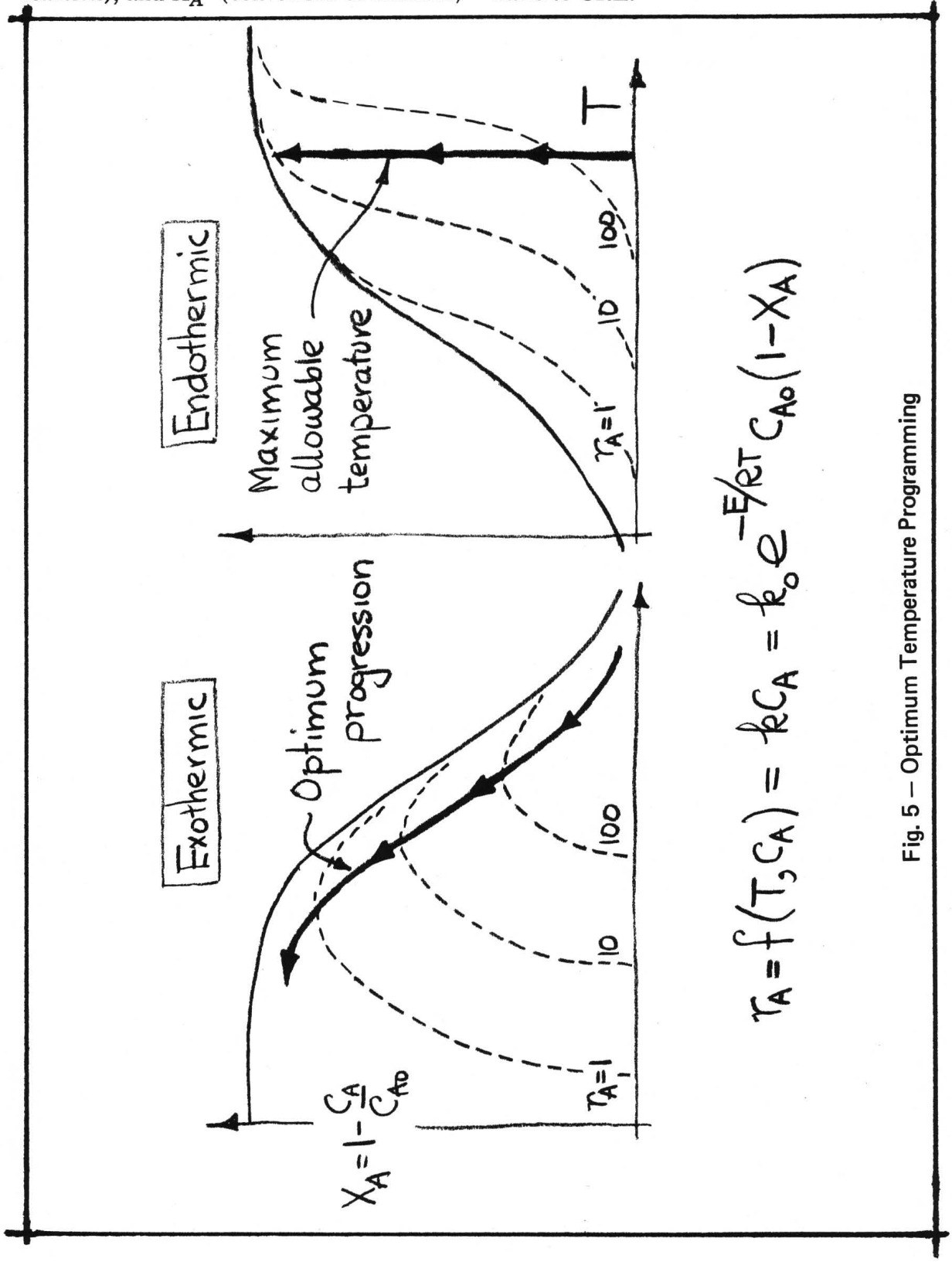


Fig. 5 — Optimum Temperature Programming

Figure 4 is the temperature-conversion graph for typical exothermic and endothermic reactions when starting with a given feed. Shown in this figure are the equilibrium curves obtained from thermodynamic considerations and typical reaction paths starting with feed at temperature T_0, both for isothermal and adiabatic operations. In the succeeding figures we shall build on this plot. I shall discuss only the exothermic situation, but I include in the figures the corresponding endothermic reaction for the reader who wishes to pursue that case on his own. The logic of the two situations is somewhat similar.

Now to Figure 5: the dotted lines show the locus of constant reaction rate, and this general shape again is typical of all re-

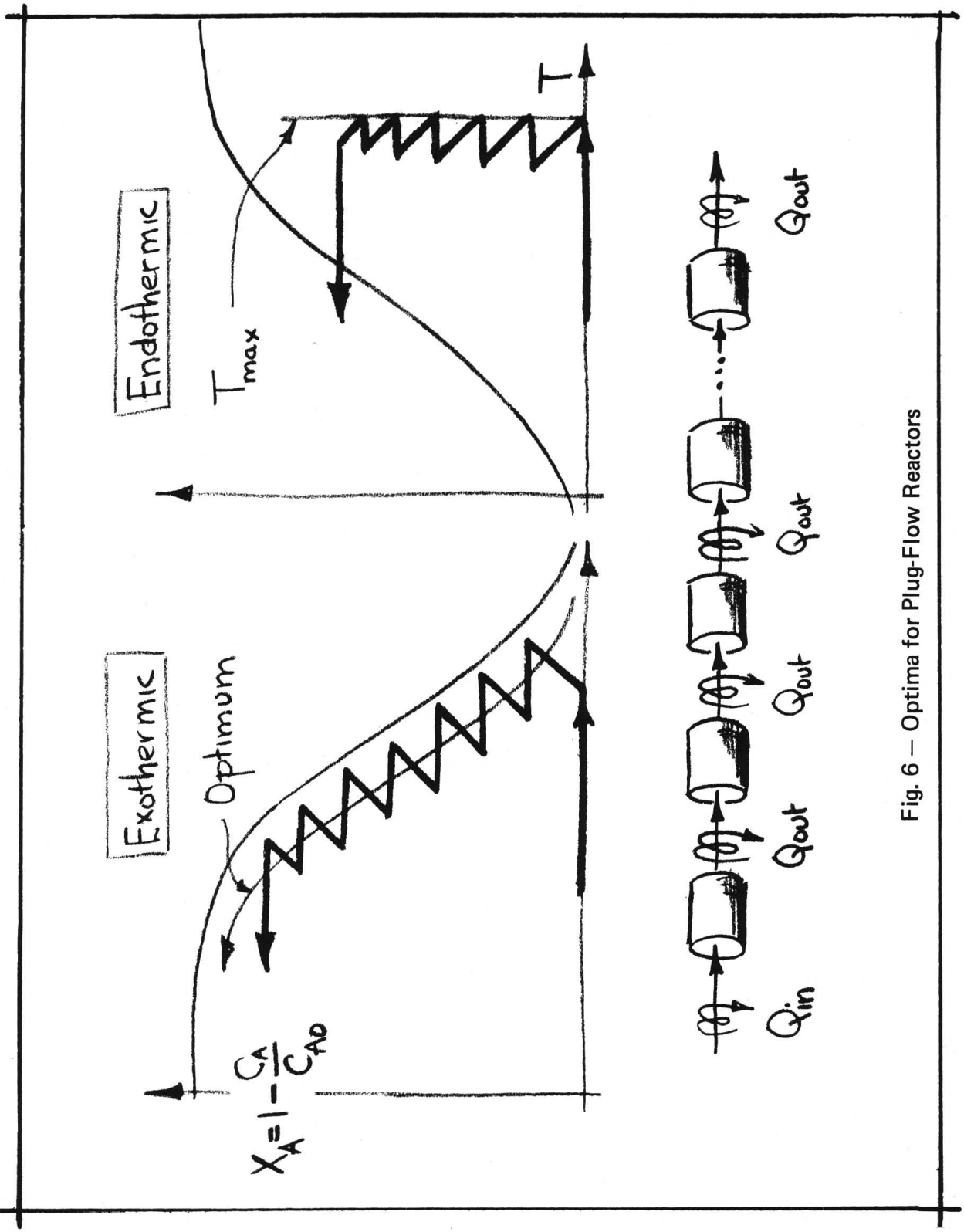


Fig. 6 — Optima for Plug-Flow Reactors

actions. Next, to minimize the size of reactor we want to choose the temperature where the rate is maximum for that composition. The heavy line in this figure shows the progression of temperatures which satisfy this condition. Comparing with Figure 4 we see that the slope of this optimum line is opposite that for adiabatic operations.

Because of the opposite slopes the way to approach this optimum with plug flow units is shown in Figure 6. Abiabatic stages with intercooling are required. It is apparent that intercooling simply shifts the operating line horizontally to the left on this graph.

Since it is impractical to use a large number of stages, we may start by examining one-stage, two-stage, and then three-stage operations. Figure 7 illustrates how

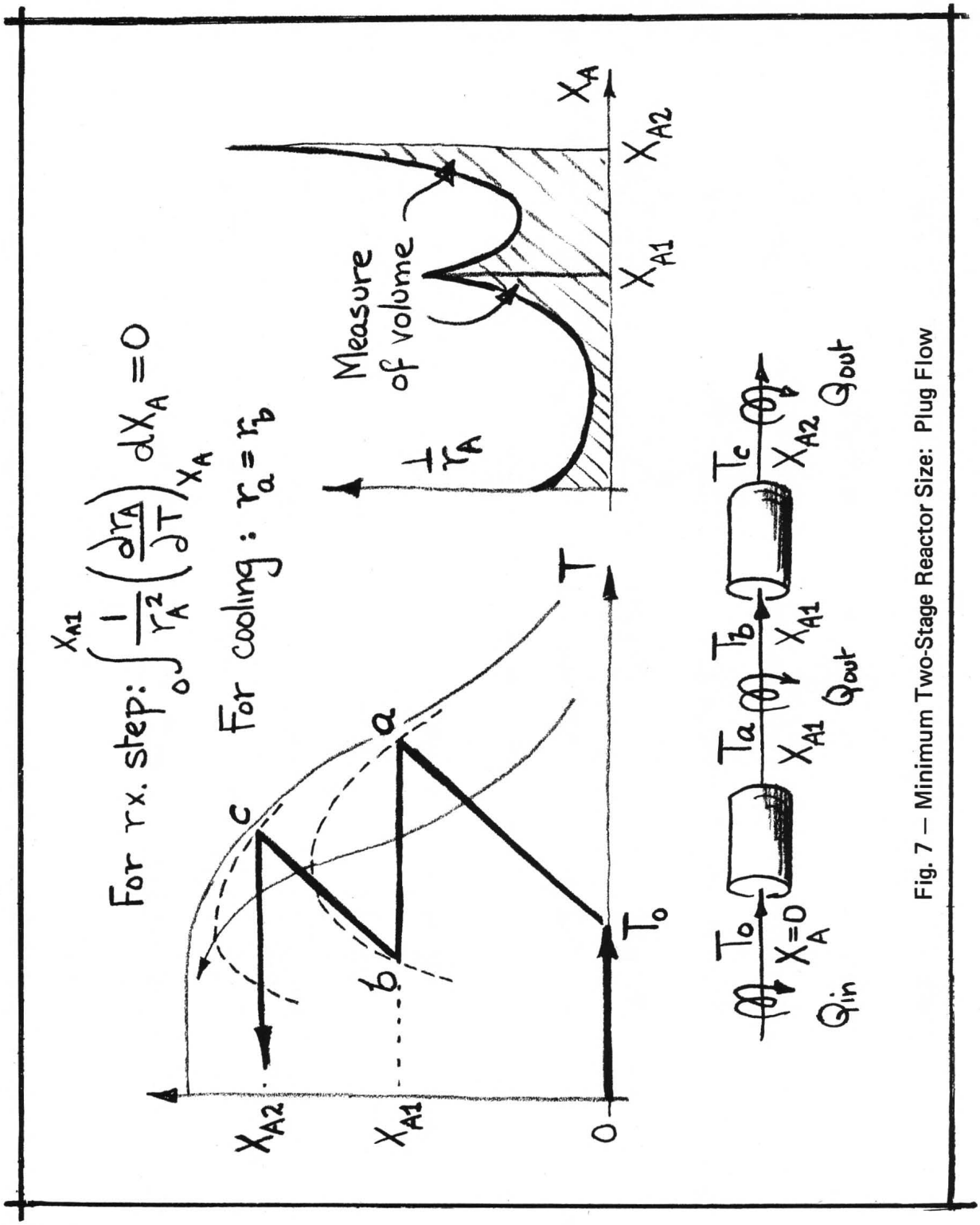


Fig. 7 — Minimum Two-Stage Reactor Size: Plug Flow

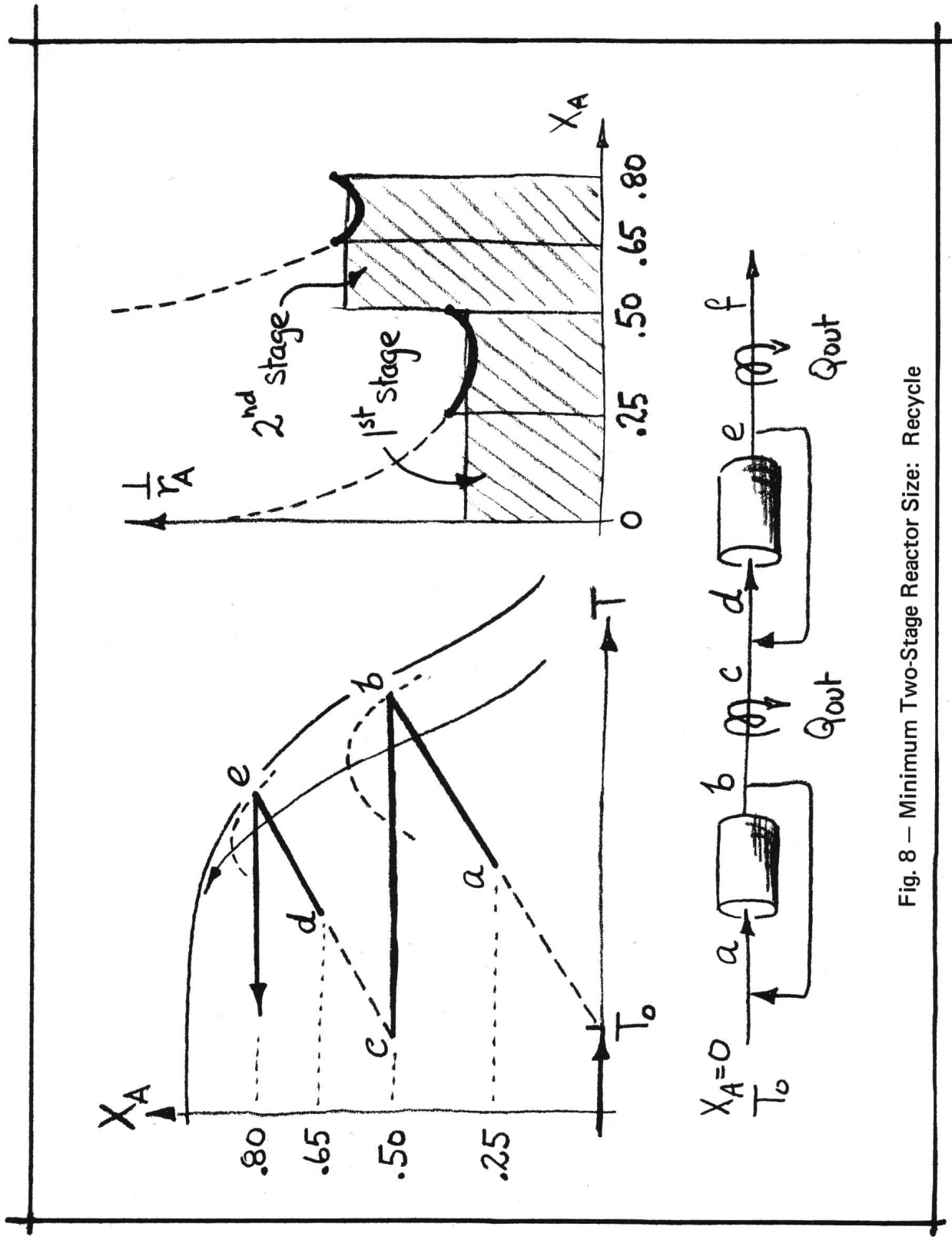


Fig. 8 — Minimum Two-Stage Reactor Size: Recycle

to find the minimum size of a two-stage plug-flow unit to achieve a given conversion XA_2. The procedure is a one dimensional trial-and-error where T_0 alone is guessed. Points *a* and *c* are found by the integral given in the figure and point *b* has the same rate as point *a*. Figures 8 and 9 sketch analogous plots for recycle and for backmix flow.

Again we ask if we must search all these possibilities. Figure 10 indicates that this is not necessary and that the key to the proper contacting is given by the slope of the adiabatic operating line. For the shallow slope of case *a* the rate of reaction is extremely low at low conversions; hence we should avoid operating in this range of conditions. This immediately suggests that we use backmix flow or recycle flow with a high

(continued on page 64)

(continued from page 62)

recycle rate. On the other hand plug flow is quite satisfactory for the steep slope of case *b*. The numerical values shown on this slide represent a heat of reaction of 30 Kcal. So, simply by making this type of graph we can tell what class of contacting schemes to examine and what class to reject.

Other alternative arrangements can also be considered, for example the cold-shot cooling of Figure 11. This arrangement eliminates all the interstage heat exchangers; however it is only practical to use this scheme under the conditions shown in the figure.

Only a detailed cost study of exchangers, catalyst, pumping, and the like will tell which set-up is best in a specific situation; however, I like this graphical procedure because it is rapid and simple to use, it is general, and it is easily extended to non-abiabatic and to homogeneous reactions. It

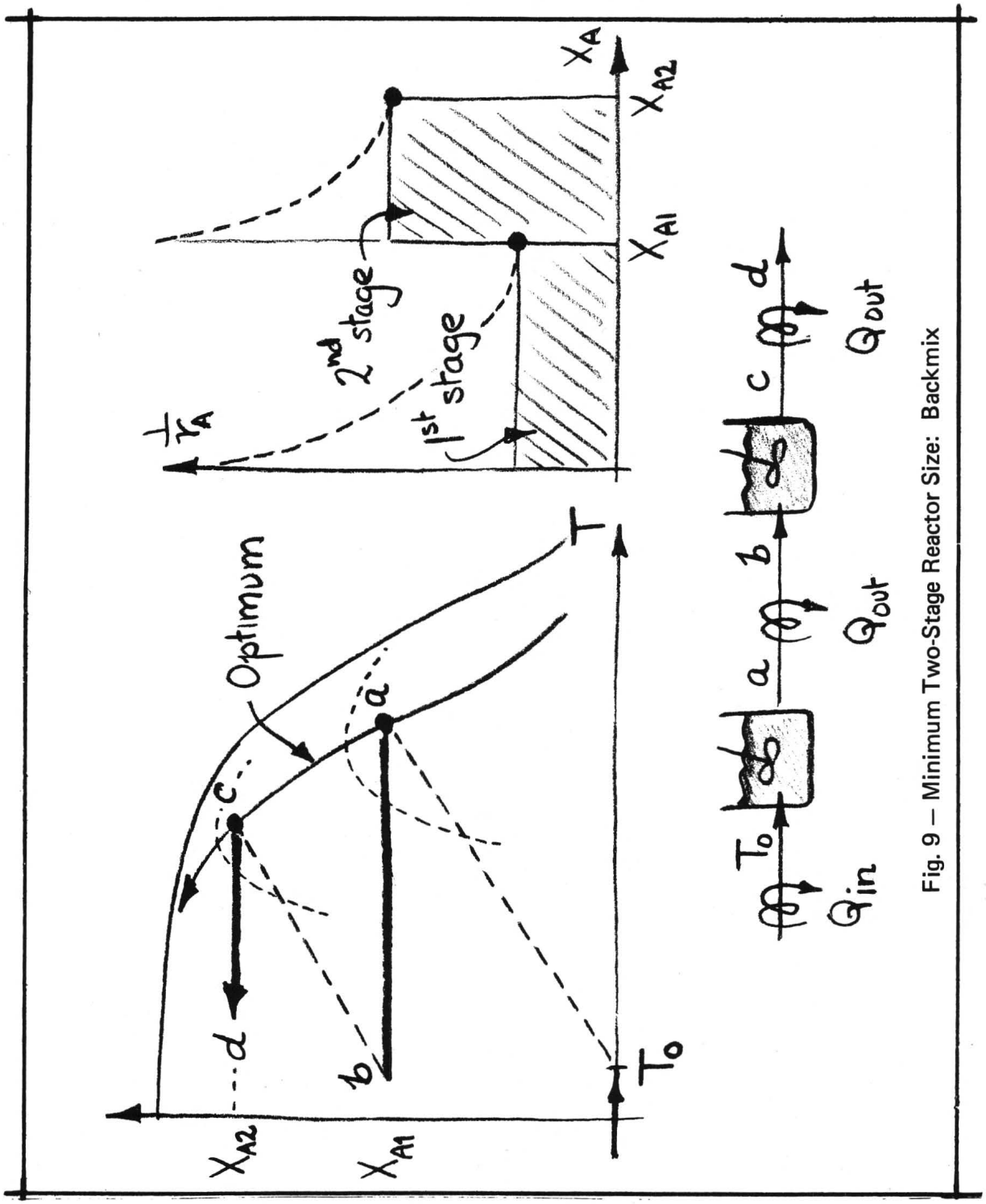


Fig. 9 — Minimum Two-Stage Reactor Size: Backmix

also gives a visualization, a feel, for what is going on. I can't help thinking that its role in reactor design is somewhat like that of the McCabe-Thiele method in distillation.

But enough. Let us look at another area where a search for generalizations is going on. This concerns multiple reactions and the question of product distribution.

Suppose we have a feed consisting of the two dichloro compounds shown on the left side of Figure 12 and we want to know how to contact this stream with chlorine so as to maximize the formation of the desired trichloro compound. Should we use countercurrent contacting, concurrent contacting, a vigorously agitated vat through which chlorine is bubbled, or some other arrangement? The direct way of answering this question is to evaluate the five rate con-

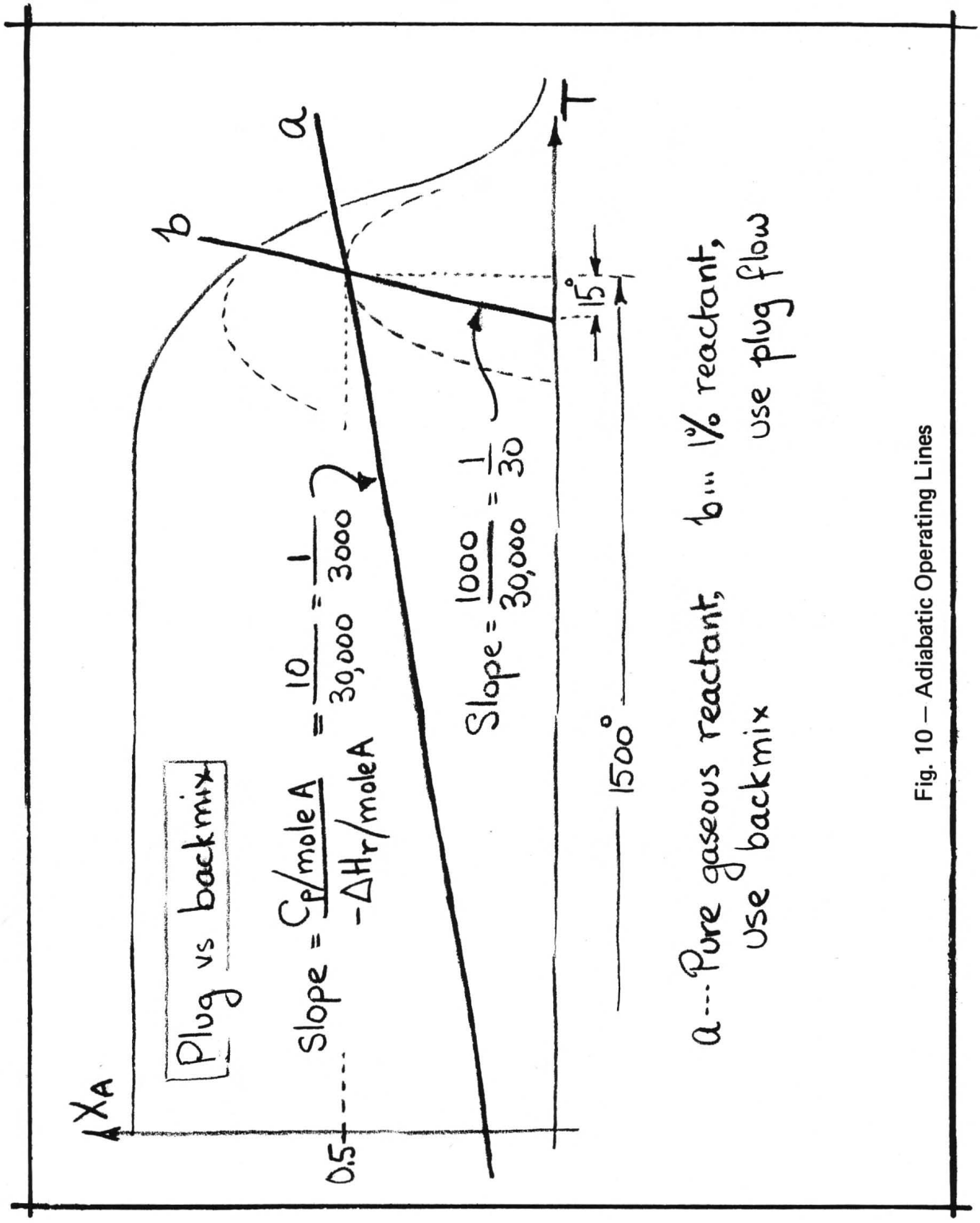


Fig. 10 — Adiabatic Operating Lines

stants and then to solve the design equations for the various contacting patterns. It would be very nice, however, if we had some general rules allowing us to find the best contacting pattern, not only for the reaction shown in Figure 12 but for other reactions as well, without going through the detailed calculations, or better still, without even knowing the values of the rate constants. Such rules would be a useful guide and a great timesaver.

A number of such rules have indeed been proposed to date. Some have been proved, while others are still conjectures. Let me present a few of them.

Rule 1. For reactions in *parallel*, for ex-

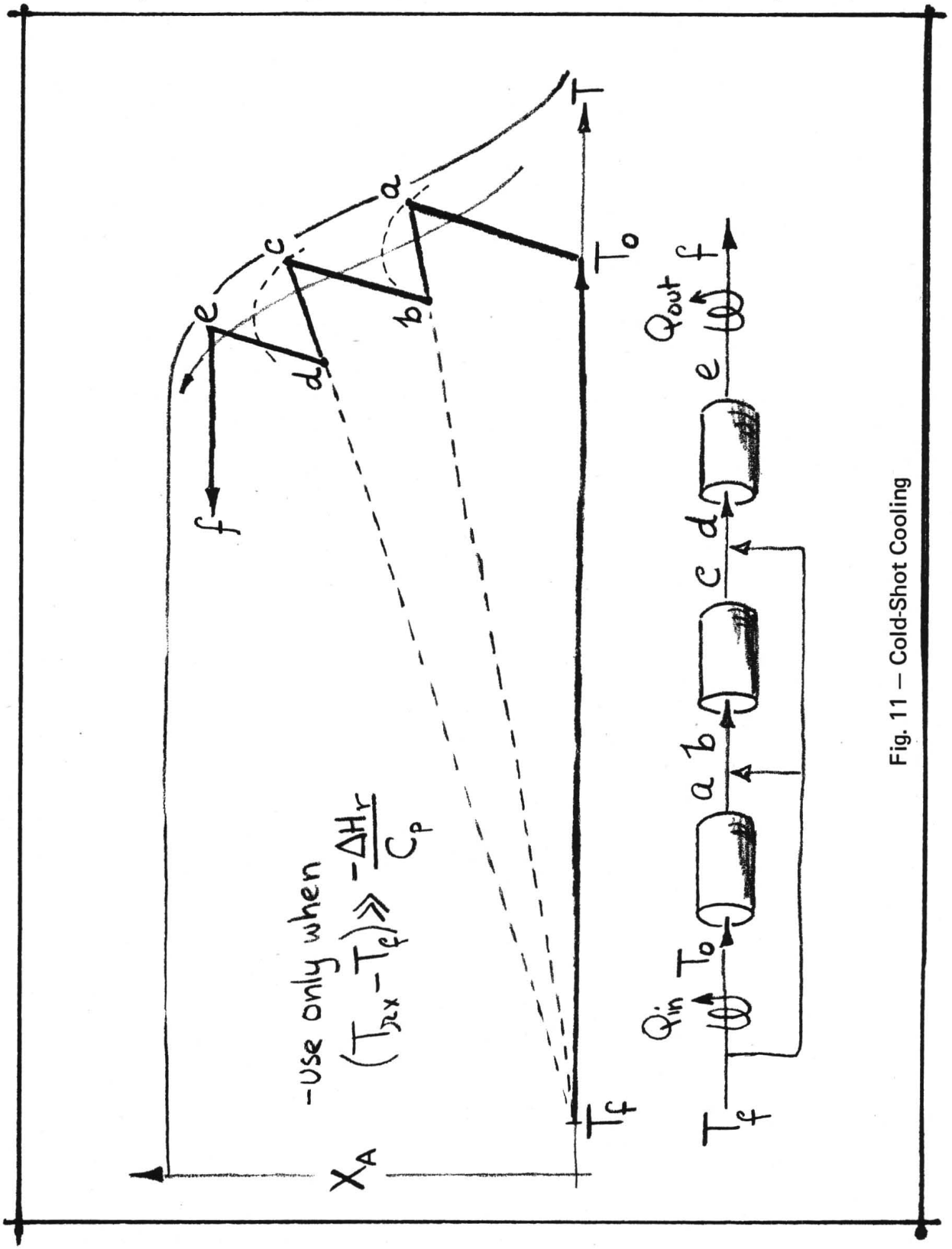


Fig. 11 — Cold-Shot Cooling

ample $A \lessgtr {R \atop S}$, the concentration level of materials is the key to proper control of product distribution: a high concentration of a component favors the reaction of higher order with respect to that component, a low concentration favors the reaction of lower order.

This rule is easy to prove for parallel reactions of all kinds including catalytic and autocatalytic.

Rule 2. For reactions in series, say $A \rightarrow R \rightarrow S \rightarrow T$, keeping the composition homogeneous, thus not allowing material of different composition to mix, will allow a maximum amount of any intermediate to be formed.

This rule has also been proved in general for any stoichiometry and reaction kinetics and for any intermediate in the reacting chain.

Rule 3. Multiple reactions can be looked upon as a combination of their constituent series and parallel reactions, as far as product distribution is concerned. For example, the consecutive competitive reaction scheme

$$A + B \rightarrow R$$

$$R + B \rightarrow S$$

can be looked upon as the sum of

$$A \rightarrow R \rightarrow S \quad \text{and} \quad B \lessgtr {R \atop S}$$

This rule is useful and allows us to break down complex reaction schemes into their building blocks, from which the optimum contacting pattern can easily be found.

Rule 4. With respect to product distribution every non-continuous contacting setup has its steady-state continuous-flow analog and vice versa.

This rule suggests, for example, that if you can produce a polymer with a specific molecular weight distribution in batch operations then you should be able to design a

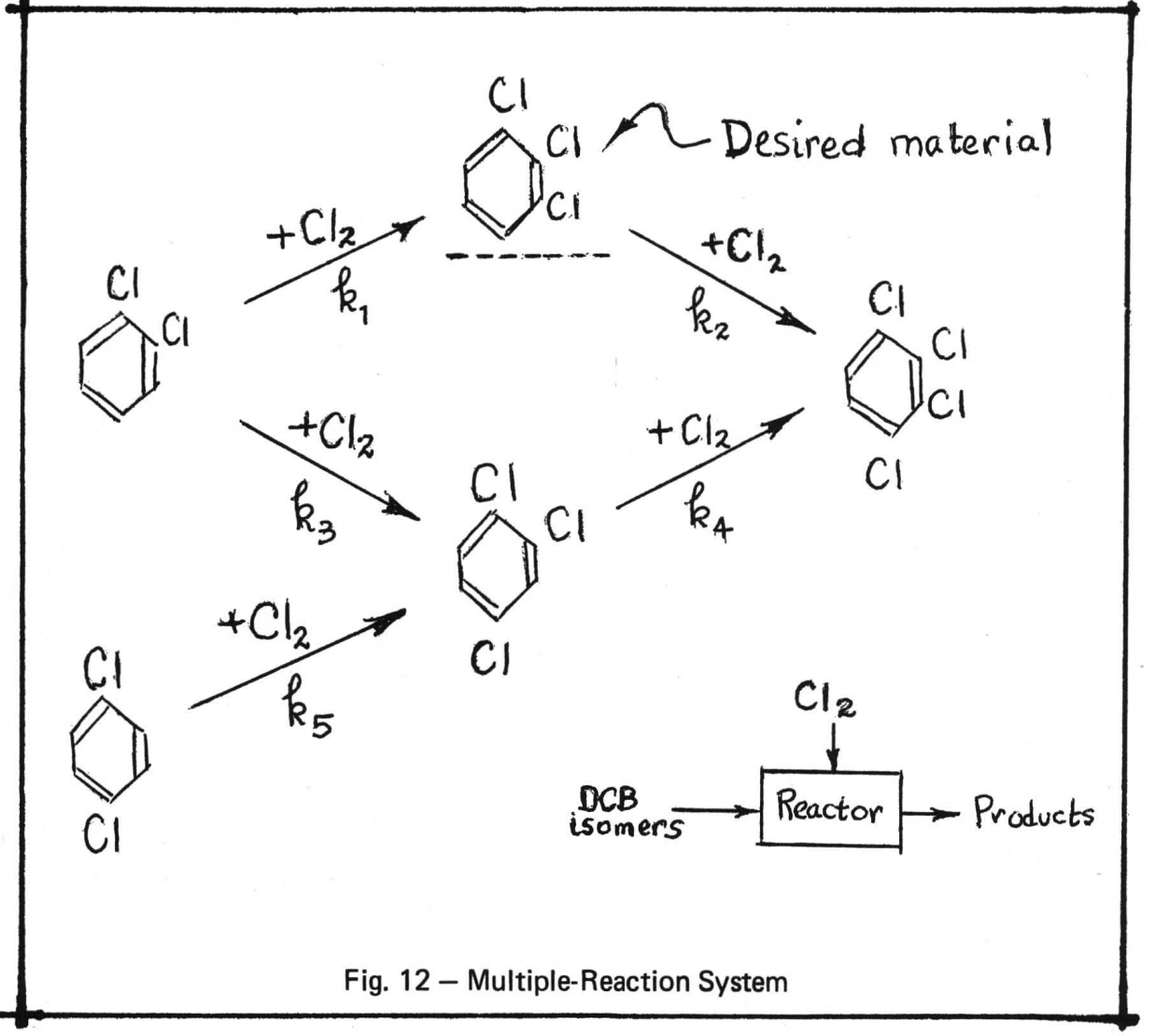
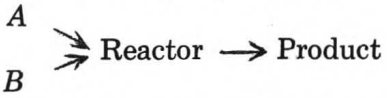


Fig. 12 – Multiple-Reaction System

continuous process which will give the same quality product. Also beer, traditionally batch-prepared, should be produceable continuously*. I suggest this rule with some hesitation because it has occasioned rather antagonistic responses, with counterexamples quoted from the literature. In fact, none of the counterexamples have held up. Thus, the rule is still open to question — it has yet to be proved or disproved.

Rule 5. For a given set of reactions, optimum contacting is independent of the value of the rate constants.

This rule certainly holds for some of the simpler reaction schemes, such as those indicated in Rules 1, 2, and 3. It can be shown by counterexample, however, that it does not hold for all reaction schemes. My opinion is that it holds if reactants enter in separate feed streams and if their introduction can be controlled, thus



but that it does not hold if the reactants enter or are present in the reactor in a fixed ratio which we are not able to control, say if they enter in one stream

Rule 6. The steady-state optimum can never be surpassed by unsteady-state operations.

This is an interesting rule because recently a number of papers have appeared suggesting that unsteady state operations can raise conversions and improve product distribution. All right then, in terms of general classes of reacting systems what are the limitations to this rule? Is it applicable to homogeneous systems? To those with mass-transfer-controlled kinetics? This is what we need to know.

I could suggest others, but I think that these six are enough to indicate the sort of thing we are looking for. Actually we are just beginning timidly to probe this area, uncertain even of the questions to be asked, never mind the answers. What we need is our own modern day Euclid for this area of CRE.

The examples cited have been intended to demonstrate that the concern and emphasis in CRE is quite different from the approaches used in the past. Also, with all the discussion in educational circles about what constitutes proper research, I cannot escape the conclusion that this type of activity, this search for generality which is useful for design and prediction, is indeed *engineering research* in the proper sense of the term.

*It is being so produced. Ed.

Now to engineering education: here we must ask how best to prepare students for prediction and design for the problems of tomorrow. Well, the basis of all experience has shown that generalizations and universals are our best bet, not the details and particulars of this or that phenomenon or process. Generalizations constitute teachable knowledge, whereas the teaching of facts that stand alone and outside of any general conceptual framework is simply the passing on of today's art. In fact, I may go as far as to say that until a general point of view is taken a subject cannot be taught effectively.

To show that CRE does constitute such teachable knowledge let us see what happened to the educational pattern after CRE appeared on the scene. First of all the first congress on CRE, where its program was first spelled out, was held in 1957. The 1962 ASEE survey of under-graduate chemical engineering education in the US gave these figures:

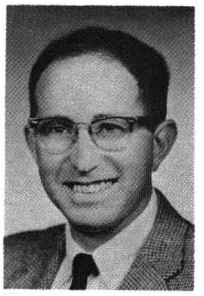
In 1957 18.5% of schools taught "Kinetics"
In 1961 53.2% of schools taught "Kinetics"

Professor Thatcher, who prepared this report, considered this large jump to be the most significant shift in the chemical engineering curriculum in this period. Why the increase? Is it a coincidence that not long after the CRE approach was introduced this subject — which had always been important — all of a sudden became teachable? I'm convinced that it is no coincidence. And if you compare the subject matter — before and after — in these courses I'm confident that you too will say that the CRE approach was largely responsible for this shift.

As of today my guess is that the 53% figure has risen to over 80%. This is a remarkable development especially in view of the fact, as I mentioned earlier, that it has taken place without major discussion in ASEE or AIChE.

Finally I'd like to say a few words about CRE and the main line evolution of the profession. Early in this century the unit operations approach was the unifying principle which brought order to the study of the physical operations of chemical technology, and it became the focal point about which chemical engineering as a profession achieved its identity, flourished, and grew. Now, fifty years later, in the midst of much questioning in the profession about identity and purpose, I see the promise of a similar but broader unification, this time encompassing the chemical operations and with CRE as its vehicle. Such is the role in our ever-changing profession which I envision for CRE, an approach whose name and program were clearly spelled out a short nine years ago. Only time will tell whether we will shape CRE to fulfill this promise.

Octave Levenspiel was born in 1926 in Shanghai, China, and grew to manhood there. He came to the United States in 1946, became a citizen in 1958, is now married and has three children.



He attended a German elementary school, British secondary school and French University (Universite l'Aurore) all in Shanghai. Dr. Levenspiel received his B.S. degree in chemistry in 1947 from the University of California at Berkeley and his M.S. and Ph.D. degrees in chemical engineering in 1949 and 1952 from Oregon State University.

Except for one year in research at University of California (1951-52), Dr. Levenspiel has been continuously in engineering education. He taught at Oregon State University (1952-54), Bucknell University (1954-58) and Illinois Institute of Technology (1958 to the present), where he is now Professor of Chemical Engineering. In 1963-64 he spent a year at Cambridge University as a National Science Foundation Senior Post-doctoral Fellow.

In research and scholarly activities his interests have centered about the use of knowledge and the development of the general approach for prediction and design. In this line he has published articles on heat transfer, fluidization, the design of chemical reactors and flow patterns of fluids in vessels. He has written a review on the last named subject in "Advances in Chemical Engineering", Vol. IV. In 1962 he wrote "Chemical Reaction Engineering" to introduce students to the method of design of chemical reactors.

Dr. Levenspiel is a Professional Engineer (State of Oregon) and has strong interests in the philosophy and history of science, and in statistics.

(continued from page 63)

of his own academic research. The student will inherit a long overdue legacy if administration in the engineering colleges will recognize that the intangible non-research activity so necessary for analysis, synthesis, and design is not only academically legitimate, but important.

Many industrial managers hold the view that the market for highly theoretical and research trained engineering Ph.D.'s in narrow specialty areas is rapidly becoming saturated. Progressive graduate schools might very well either: 1, consider adoption of a requirement that a portion of the engineering doctoral candidates demonstrate an acceptable degree of competence in design, or 2, adopt curricula which will produce broadly trained design professionals at the advanced degree level who are able to utilize modern computational facilities to the fullest extent. Training of our young people cannot be done by professors who are themselves design illiterates.

Solutions to the problem of design instruction will not fall out easily. We do, however, have an effective way of using a new dimension in industry-university cooperation to scaledown the problem.