

TEACHING REACTION ENGINEERING USING THE ATTAINABLE REGION

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Ask a graduating chemical engineering student the following question: What makes one reactor different from the next? The answers received will often be unsatisfactory and vary widely in scope. Some may cite the difference between the basic design equations, others may point out a PFR is “longer,” and still others may state that it all depends on the particular reaction network. Though these answers do possess a bit of truth, they do not capture the true difference between reactors: the degree of mixing achieved. This is the inherent difficulty with teaching chemical reaction engineering. The students learn the technical skills required to perform the calculations to determine maximum yields and shortest space-times, but very rarely are they able to grasp and thoroughly understand the theory and underlying differences between reactors.^[1] Often, too much time is devoted to tedious and involved calculations to determine the correct answer on homework instead of focusing on the concepts to enforce the benefits offered by each reactor presented.

Reactor network optimization is traditionally not covered in any depth at the undergraduate level.^[2-4] The way reactor network optimization is traditionally taught to graduate students often involves large numbers of coupled equations that can sometimes hide the final goal of the analysis. Attempts

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to simplify the situation, such as Levenspiel's graphical analysis,^[4] do offer some benefit, however their applicability is limited as they can readily only optimize simple reaction problems. For chemical engineers, it is paramount to know the most promising solution to a real problem in the shortest amount of time, and rarely is this accomplished with the current teaching methods for reactor network optimization.

Presented here is an approach that addresses the challenges presented above. The attainable region (AR) approach is a powerful research technique that has been applied to optimization of reactor networks.^[5-7] It is also a powerful teaching tool that focuses on the fundamental processes involved in a system, rather than the unit operations themselves. It has been used to introduce undergraduate and graduate students to complex reactor network optimization in a short time, with little to no additional calculations required.

BACKGROUND

The generic approach to complex reactor design and optimization is to build on previous experience and knowledge to test a new reactor configuration against the previous champion that yielded the best result.^[8] If a new maximum is achieved, the reactor configuration and process settings are kept. If not, the previous solution is retained and the entire process is repeated. The biggest issue with this trial and error approach is the time it takes. Also, there is no way to know if all possible combinations of operational parameters and reactors have been tested. In addition, calculations are normally exhausting and general computational techniques are difficult to develop due to the specificity of each arrangement. Over time, this mechanism has evolved into a set of design heuristics that dominate decision processes throughout industry.^[9]

Achenie and Biegler^[10] model a reactor superstructure using a mixed integer nonlinear programming (MINLP), which transforms the task into an optimal control problem. Again, this approach is useful if the optimal reactor network is known, but it does not address the issue of choosing the optimal reactor network.

Horn^[11] defines the AR as the region in the stoichiometric subspace that could be reached by any possible reactor system. Furthermore, if any point in this subspace were used as the feed to another system of reactors, the output from this system would also exist within the same AR. This framework approaches reactor design and optimization in a simpler, easier, and more robust manner. It offers a systematic *a priori* approach to determining the ideal reactor configuration based upon identifying all possible output concentrations from all

possible reactor configurations. One of its advantages over previous approaches is the elimination of laborious and counter-productive trial and error calculations. The focus is on determining all possible outlet concentrations, regardless of the reactor configuration, rather than on examining a single concentration from a single reactor. Approaching the problem from this direction ensures that all reactor systems are included in the analysis, removing the reliance on the user's imagination to create reactor structures. Also, for lower dimensional problems often studied in the undergraduate courses, the final solution can be represented in a clear and intuitive graphical form. From this graphical representation, the optimal process flow sheet can be read directly. In addition, once the universal region of attainable concentrations is known, applying new objective functions on the reactor system is effortless. No further calculations are required, and the optimal values can be read directly from the graph.

Finally, this general tool can be applied to any problem whose basic operation can be broken down into fundamental processes, including isothermal and nonisothermal reactor network synthesis,^[5,12] optimal control,^[13] combined reaction and separation,^[14-16] comminution,^[17,18] and others. Process synthesis and design usefulness are aided greatly by this alternative approach.

The AR analysis method has been presented in undergraduate courses, to industrial audiences, and in master's courses at the University of the Witwatersrand in South Africa, as well as, more recently, as an alternative to traditional complex reactor design in a graduate reaction engineering course at Rutgers University. The overall response from the audiences has been favorable, and it is the intention of the authors to discuss the benefits this approach offers to the field of reaction engineering. To aid with teaching/learning, a detailed attainable region Web site has been set up and the address is given at the end of this article.

In this paper we will first introduce a moderately challenging reaction engineering problem. Next, the AR analysis will be illustrated by solving the presented problem. Finally, the teaching strategy adopted by both institutions will be presented.

PROBLEM STATEMENT

The following liquid phase, constant density, isothermal reaction network will be used to illustrate the AR approach.



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The initial characteristics of the reaction network are shown in Table 1. The end goal of this exercise is to determine the reactor configuration that maximizes the production of B for

C_A^0	C_B^0	C_C^0	C_D^0
1 kmol m ⁻³	0 kmol m ⁻³	0 kmol m ⁻³	0 kmol m ⁻³
k_1	k_2	k_3	k_4
0.01 s ⁻¹	5 s ⁻¹	10 s ⁻¹	100 m ³ kmol ⁻¹ s ⁻¹

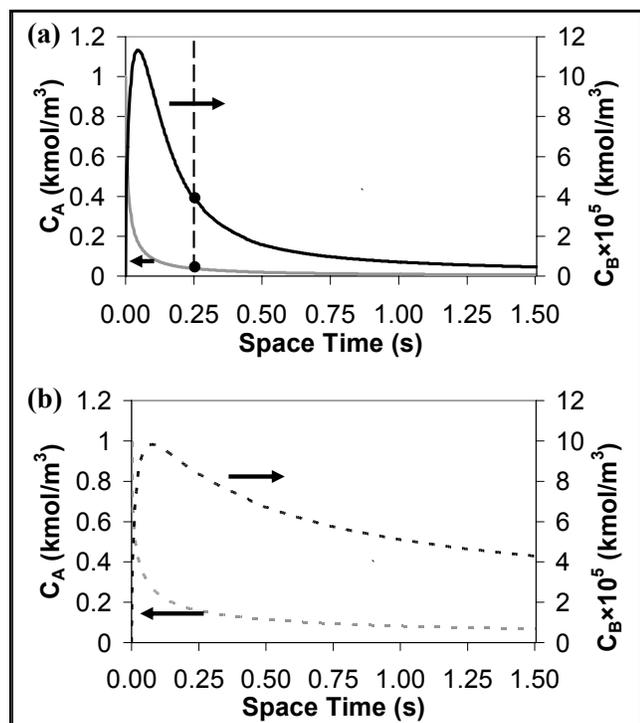


Figure 1. Concentration as a function of space-time in a PFR (a) and CSTR (b). Note that profiles for C_C and C_D are not shown.

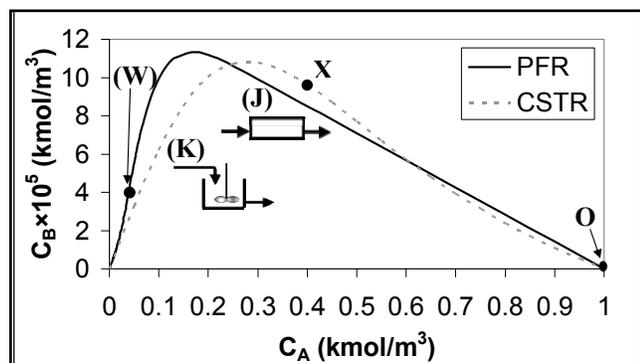


Figure 2. State-Space diagram. Point O represents the feed point. Point X represents an arbitrary CSTR effluent point. The diagram on the top right is a PFR representing the PFR profile, (J). The diagram in the bottom left is a CSTR representing the CSTR locus, (K).

a feed of pure A. These reaction kinetics were used because they represent a reaction network without an intuitively obvious optimal structure. A PFR will maximize the amount of B produced in the first reaction, but a CSTR will minimize the amount of A consumed in the second reaction.

SOLUTION

Determining the candidate attainable region for this reaction scheme involves the completion of the following simplified steps: selecting the fundamental processes, choosing the state variables, defining and drawing the process vectors, constructing the region, interpreting the boundary as the process flow sheet, and finding the optimum.

I. Choose the Fundamental Processes

In this particular example, the fundamental processes are reaction and mixing. Let us first look at mixing. There are two limits on mixing in a reactor: a plug flow reactor, in which a slug of fluid does not experience any axial mixing along the reactor length, and a continuously stirred tank reactor, in which each volume element experiences complete mixing. Before moving further into the analysis, it is useful to determine the dependence of species concentrations on space-time in these two environments. For a PFR, this is determined by numerically solving the mass balances in Eqs. (3)-(6), giving the concentration profiles of C_A and C_B in Fig. 1(a).

$$\frac{dC_A}{d\tau} = -k_1 C_A + k_2 C_B - k_4 C_A^2 \quad (3)$$

$$\frac{dC_B}{d\tau} = k_1 C_A - k_2 C_B - k_3 C_B \quad (4)$$

$$\frac{dC_C}{d\tau} = k_3 C_B \quad (5)$$

$$\frac{dC_D}{d\tau} = k_4 C_A^2 \quad (6)$$

Similarly, the set of mass balances in Eqs. (7)-(10) can be solved to give the locus for a CSTR as τ is varied, provided in Fig. 1(b).

$$C_A - C_A^0 = \tau(-k_1 C_A + k_2 C_B - k_4 C_A^2) \quad (7)$$

$$C_B - C_B^0 = \tau(k_1 C_A - k_2 C_B - k_3 C_B) \quad (8)$$

$$C_C - C_C^0 = \tau(k_3 C_B) \quad (9)$$

$$C_D - C_D^0 = \tau(k_4 C_A^2) \quad (10)$$

In Eqs. (3)-(10), C_i represents the concentration of species i , C_i^0 represents the feed concentration of species i , τ is the space-time of the reactor, and k_j represents the rate of reaction j . Figure 1 only shows the profiles for C_A and C_B because, as will be explained shortly, C_C and C_D do not influence the determination of the AR.

II. Choose the State Variables

The state variables for this example are C_A and C_B . C_B is a state variable because it is the value that we wish to optimize.

C_A is a state variable because, looking at the right-hand side of Eqs. (3)-(10), the behavior of C_B is entirely dependent on the change in C_A . Note that τ is not a state variable because it is the independent variable in the system.

Now that the state variables are known, a state-space or phase-space diagram^[19] (Figure 2) can be created showing the autonomous relation between C_A and C_B . First, we must do this for the PFR using the data in Figure 1(a). Figure 1(a) shows C_A and C_B as a function of τ , so for any given τ we can determine a C_A, C_B pair, which allows us to plot curve (J) (solid line) in Figure 2. For example, the point W in Figure 2 corresponds to $C_A = 3.81 \times 10^{-2}$ kmol/m³ and $C_B = 3.95 \times 10^{-5}$ kmol/m³, and can be traced back to $\tau = 0.25$ seconds in Figure 1(a). The same can be done for the data in Figure 1(b) for the CSTR that leads to curve (K) (dashed line) in Figure 2. While space-time is not explicitly shown in Figure 2, the relevant space-time to achieve a given concentration can always be obtained from Figure 1 (or an equivalent figure). A candidate for the attainable region (AR^C) is identified as the union of the regions contained under both curves.

III. Define and Draw the Process Vectors

A process vector gives the instantaneous change in system state caused by that fundamental process occurring. For example, if only reaction is occurring, the reaction vector, $r[C_A, C_B]$, will give the instantaneous direction and magnitude of change from the current concentration position. For mixing, this vector gives the divergence from the current state, c , based upon the added state, c^* , or $v(c, c^*) = c^* - c$. T is some arbitrary effluent concentration from a CSTR shown strictly for demonstration purposes.

The vectors can be graphically represented by considering curve (K) for the CSTR in Figure 2. This is replotted in Figure 3 along with the direction of each rate vector. The CSTR rate vector (OT) is co-linear with the feed and effluent concentrations, and the mixing vector (OX) linearly connects the current state with the added state. The resulting mixed state lies on the mixing line and its position can be determined from the Lever Arm Rule. One can also consider a PFR rate vector which originates at the current concentration and is tangent to the curve (see Figure 3).

IV. Constructing the Region

To construct the region, the process vector guidelines from the previous step are applied to the state-space diagram (Figure 2). The idea is to draw process vectors to extend the current AR^C. We begin the analysis by examining mixing.

Starting at a generic point X on the boundary of curve (K) in Figure 2, a straight line can be drawn to point O, which is the feed point. This is shown by line (L) in Figure 4(a). To achieve any concentration along line (L), you can mix the outlet of a CSTR operating at point X with the feed at point O. Thus, any point on curve L corresponds to a CSTR with bypass. The Lever Arm Rule^[20] can be used to determine the

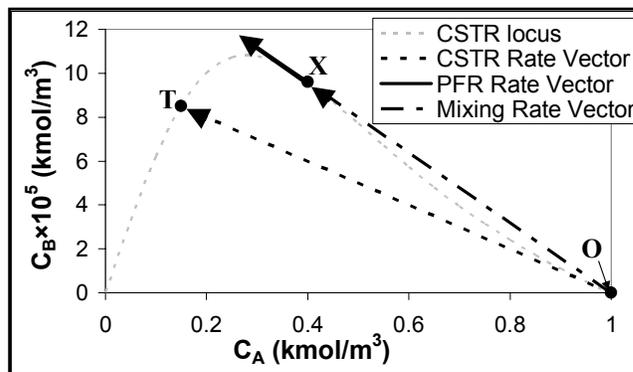


Figure 3. Rate vectors of the fundamental processes involved in the example. The CSTR rate vector points from the feed point, O, to the particular effluent point, T. The PFR rate vector is tangent to the current concentration. The mixing rate vector is a straight line pointing from the current state to the added state.

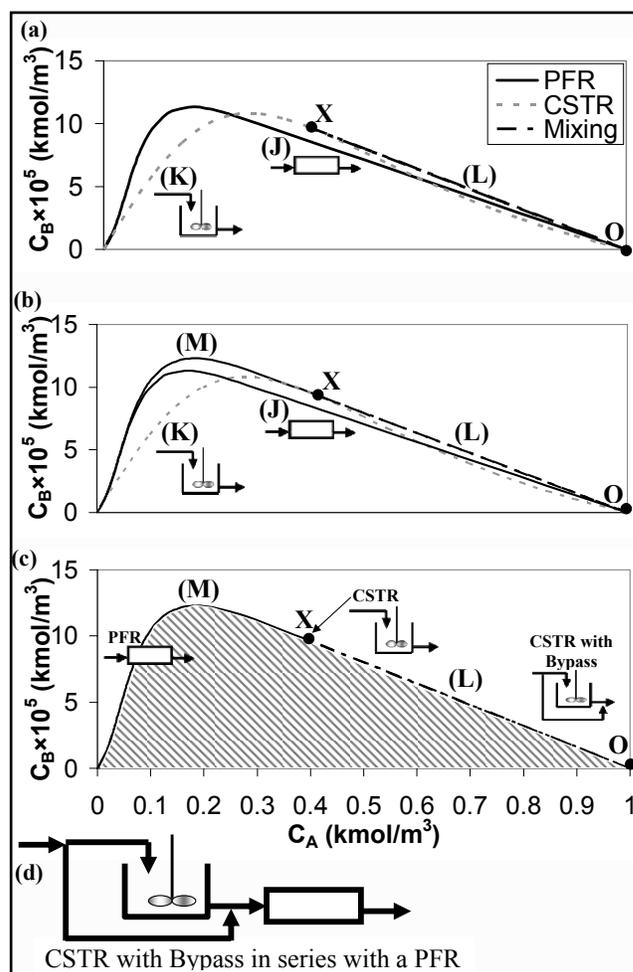


Figure 4. Determination of the Attainable Region. (a) Extension through mixing (dashed line); (b) Extend with PFR in series [curve (M)]; (c) Resulting attainable region (shaded) with corresponding reactors. Note that (a)-(c) have an equivalent x-axis. (d) Reactor configuration to achieve any point within the attainable region in (c).

percentage of each stream to mix to obtain the desired concentration. Notice that when this line is drawn, the candidate region is extended. When two states mix linearly, mixing can extend any concave region by creating its convex hull.

Does operation in a PFR extend the region as well? The answer is yes. Going back to process vector geometry, the PFR process vector is tangent to the current system-state. A line tangent to the curve at point X extends the region above its previous maximum. The complete successive PFR profile [curve (M) in Figure 4(b)] is found by numerically solving the differential PFR balance equations in Eqs. (3)–(6) with feed concentration of $X = (C_A, C_B)$. The boundary of the current candidate attainable region is now made up of curves (L) and (M) (see Figure 4b).

The attainable region can be constructed once it has been determined that no other processes can extend the region. The shaded region of Figure 4(c) shows the entire AR for this particular reaction network. The boundary of the shaded region is made up of curves (L) and (M). Since the region is convex, it is clear that mixing cannot extend the region. Moreover, it is possible to show that all rate vectors on the boundary are either tangent to the boundary or point into the region (see AR Web site for further details). Enclosed beneath the boundary are all possible reactor effluents given a feed point at O.

V. Interpret the Boundary as the Process Flow Sheet

The process flow sheet is determined by tracing a path to the point of interest. The effluent concentration at point X is achieved in a CSTR. If the desired effluent is to the right of point X on the boundary [given by curve (L)], a CSTR operating at point X with feed bypass is used to reach the point (see section III). If the desired effluent is on the boundary to the left of point X [given by curve (M)], a CSTR operating at point X followed by a PFR in series is required. These configurations are pointed out in Figure 4(c). The reactor configuration in Figure 4(d) can be used to achieve any point on the boundary of the AR^C for this reaction network.

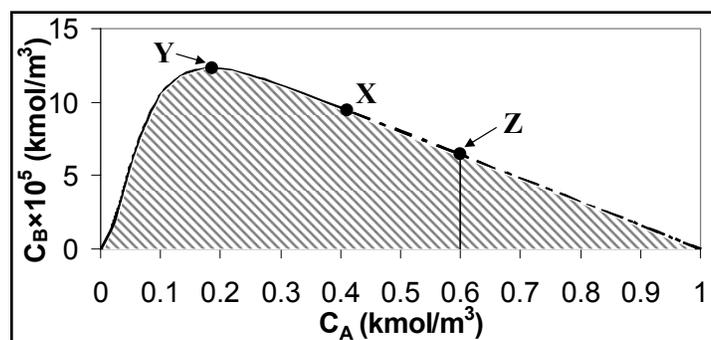


Figure 5. Application of constraints on the attainable region. Point Y: maximum B produced in reaction network. Point Z: maximum B produced given that C_A must be greater than 0.6 kmol/m^3 .

VI. Find the Optimum

The final step is to determine the optimum for the specified objective function. In this case, the objective function is to maximize the production of species B given the feed of 1 kmol/m^3 of A. It can easily be seen from Figure 5 (point Y) that a maximum of $1.24 \times 10^{-4} \text{ kmol/m}^3$ of species B can be achieved using a CSTR with effluent of 0.4 kmol/m^3 of species A followed by a PFR with an effluent concentration of A of 0.18 kmol/m^3 . The corresponding space-times of the CSTR and the PFR are 0.037 s and 0.031 s , respectively. These were determined from Eqs. (3) and (4) for the CSTR and (7) and (8) for the PFR.

With the attainable region fully determined, the optimal value for any objective function may be determined. For example, a plant manager dictates that the concentration of A cannot drop below 0.6 kmol/m^3 , or the acidity will corrode downstream equipment. The maximum amount of species B that can be produced with this constraint is given by point Z in Figure 5, which corresponds to $6.4 \times 10^{-5} \text{ kmol/m}^3$ of B. The reactor configuration that gives this outlet concentration is a CSTR with feed bypass. Cost, partial pressure, temperature, and residence time are some other examples for possible objective functions. As stated at the outset of this section, these steps are a simplified version of the rigorous procedure (see Reference 5 for more details). A final point of note is the AR analysis does not guarantee the determination of the complete attainable region. The analysis is composed of guidelines for the creation of a candidate attainable region, as no mathematically derived sufficiency conditions exist. This is the reason for the AR^C terminology.^[21]

TEACHING STRATEGY AND STUDENT FEEDBACK

At the School of Chemical and Metallurgical Engineering at the University of the Witwatersrand in Johannesburg, South Africa, the AR is taught at both the undergraduate and master's level. The AR is presented as a supplementary topic in the undergraduate Reactor Design course for third- and fourth-year chemical engineering students. After the students have developed PFR profiles and CSTR loci for a given feed concentration and reaction network, the "rules" are explained (*i.e.*, PFR rate vectors are tangent to the profile, the region can be made convex through mixing, etc.). The students are then challenged to find the region of optimal production for a certain component, and are provided with PFR profiles for various feed concentrations. At this point, the instructor emphasizes that the geometric solution the students are creating is essentially solving the same equations the students were laboring through earlier in the course. The lecturer then introduces some more complex problems involving heat transfer and reaction to demonstrate to the students the power of the method.

The AR is also taught in a week-long, 30-hour, Reactor

Synthesis Masters of Science course. The class is composed of people from industry and students who have just graduated. Therefore, the best teaching approach does not include intimidating differential equations or tedious calculations. First, the students work through the example presented in this paper as an introduction to the AR approach. Then the students are given PFR state-space profiles for different feed concentrations and asked to determine the optimal reactor configuration to achieve the maximum production of a certain species.

More recently, the AR was taught to a graduate core Reaction Engineering course of approximately 20 students at Rutgers University. Half of the students were full-time graduate students and the other half were part-time professionals who had been out of school for varying intervals. One three-hour lecture on the example covered in this paper was given after single reactor design, complex kinetics, and nonisothermal reactions had been introduced, but before biological reactions and catalysis. The technique was presented as an alternative to the computer-intensive MINLP.

Following the lecture, homework is assigned to allow the students to develop the AR themselves. The homework assignment covers a reaction network similar to the example presented, only it lacks the reversible part of the A to B reaction (also known as van de Vusse kinetics). The benefits of such an assignment are: to test basic reaction engineering skills (solving PFR and CSTR balances); to develop skills using computational programs such as POLYMATH, MATHCAD, or MATLAB; to discover the potential benefits of recycle, bypass, and Differential Sidestream Reactors (DSR) in reactor configurations; and to understand the benefits of a graphical approach to a normally calculation-intensive problem. Finally, the students are challenged on an exam with the in-class exercise given to the master's students in the Witwatersrand course.

We also feel that the AR approach lends itself well to senior design, especially in an environment where students are asked to come up with a flow sheet for their design project. These steps present a systematic approach to determining the optimal network for the reaction portion of their design project. The students can compare their initial proposals to this optimal target and decide if there is any benefit in improving their initial designs.

Some of the comments from the students included that the attainable region material was enjoyable, as "it was something new" and there was a desire to see "more advanced topics like the AR." Students were excited by the fact that they could solve problems and come up with optimum structures for reactions no one else had solved before, *i.e.*, the optimum solution was not available in any textbook or research article. Along those lines, students also commented that they liked the fact they were being taught material that was "hot off the presses" and had been the subject of a Ph.D. dissertation only a few years before.

A difficulty observed in introducing the AR to undergraduates was that some students struggled with solving new problems. In particular, students could follow the example that was developed in this article and compute the boundary of the AR themselves for a homework problem with the same basic structure, *i.e.*, a CSTR followed by a PFR. If the boundary of the AR was changed in a homework problem to a PFR followed by a CSTR followed by a PFR, however, then some students struggled with this. It was found that if these students went over a number of additional AR problems they could eventually master the material and generate ARs independently for new cases.

CONCLUSION

Reaction engineering is a course in which students often get bogged down with intensive calculations and lose sight of the more important, fundamental concepts. This paper presents the attainable region analysis method as a way to avoid this trap, and at the same time introduce design and optimization of complex reactor flow sheets—a more difficult and industrially relevant exercise. Contrary to traditional complex reactor design optimization, the AR approach does not require trial and error, does ensure that all reactor configurations are evaluated, and allows for easy application of various objective functions. Additionally, for lower-dimensional problems, the solution can be represented in a simple and clear graphical form.

The intention of the authors is to increase the exposure of this technique so that its advantages for both teaching and research can be known throughout the engineering community. The applications do not end at reaction engineering, and the reader is challenged to find areas of study to which this approach does not apply.

For more details on the attainable region approach please see the following Web site: <<http://www.wits.ac.za/fac/engineering/procmat/ARHomepage/frame.htm>>.

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