

Basics of Non-Ideal Flow

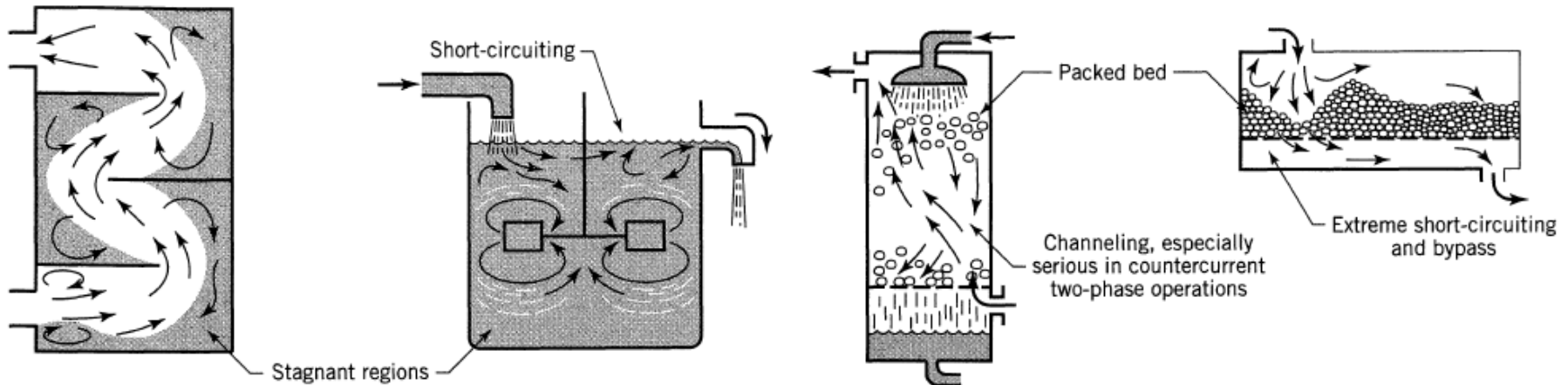
Chapter 11

- So far we have treated two flow patterns, plug flow and mixed flow.
- These can give very different behavior (size of reactor, distribution of products).
- But real equipment always deviates from these ideals. How to account for this?
- Overall three somewhat interrelated factors make up the contacting or flow pattern:
 1. the RTD or **residence time distribution** of material which is flowing through the vessel
 2. the **state of aggregation** of the flowing material, its tendency to clump and for a group of molecules to move about together
 3. the **earliness and lateness of mixing** of material in the vessel.

Let us discuss these three factors in a qualitative way at first. Then, this and the next few chapters treat these factors and show how they affect reactor behavior.

The Residence Time Distribution, RTD

- Deviation from the two ideal flow patterns can be caused by channeling of fluid, by recycling of fluid, or by creation of stagnant regions in the vessel.



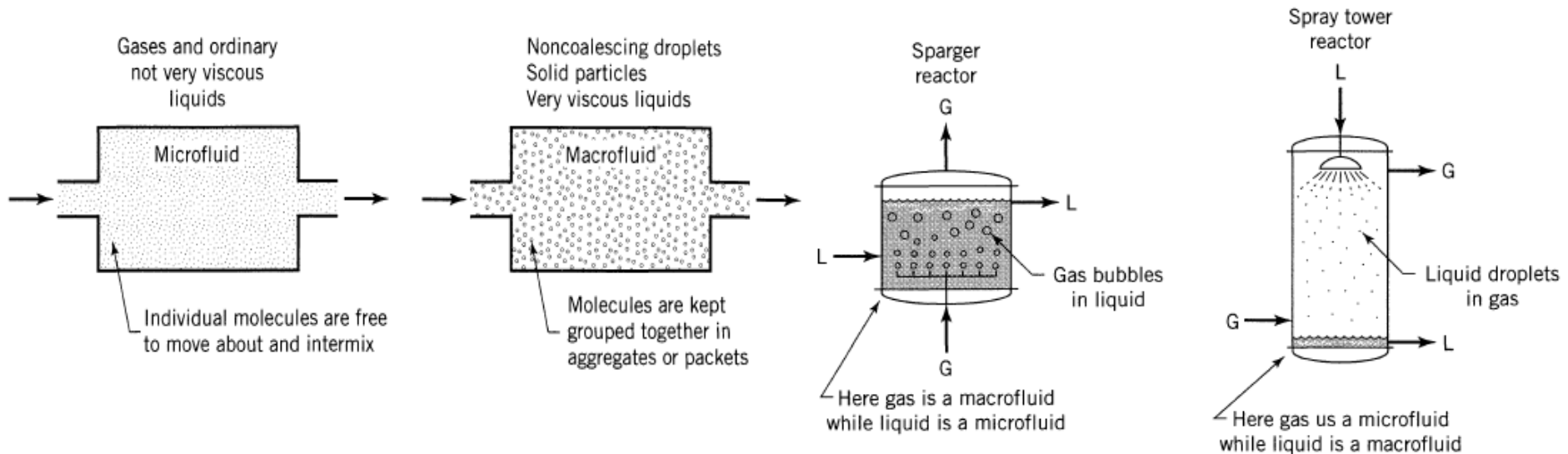
In all types of process equipment, such as heat exchangers, packed columns, and reactors, this type of flow should be avoided since it always lowers the performance of the unit.

- If we know precisely what is happening within the vessel, thus if we have a complete velocity distribution map for the fluid in the vessel, then we should, in principle, be able to predict the behavior of a vessel as a reactor.
- Unfortunately, this approach is impractical, even in today's computer age.
- In many cases we really do not need to know very much, simply how long the individual molecules stay in the vessel, or more precisely, the distribution of residence times of the flowing fluid.
- This information can be determined easily and directly by a widely used method of inquiry, the stimulus-response experiment.
- We will only consider the steady-state flow, without reaction and without density change, of a single fluid through a vessel.

State of Aggregation of the Flowing Stream

- Flowing material is in some particular state of aggregation, depending on its nature.
- In the extremes these states can be called *microfluids* and *macrofluids*,

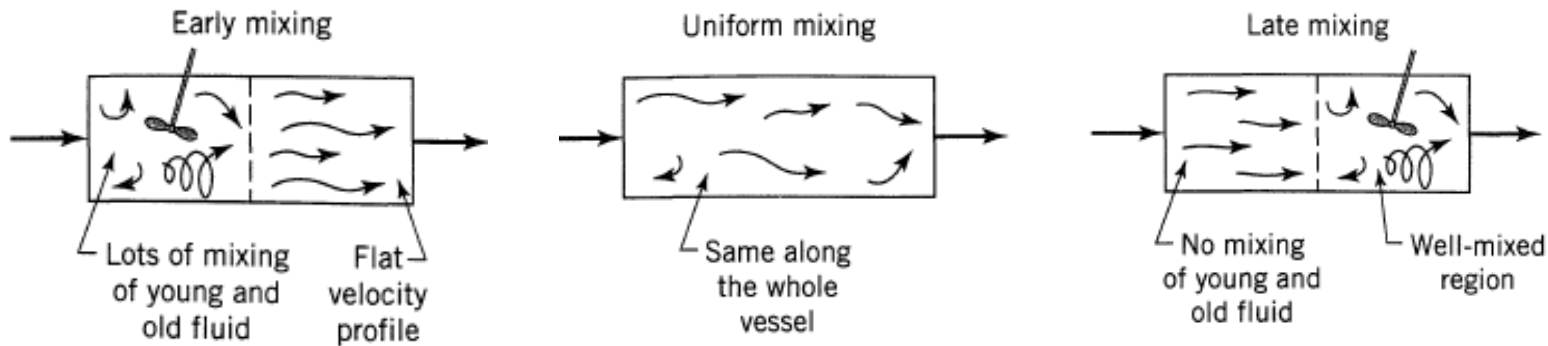
Single-Phase Systems. These lie somewhere between the extremes of macro and micro fluids.



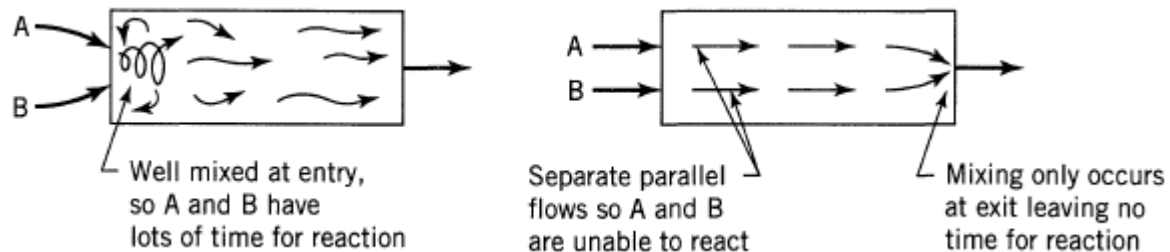
Two-Phase Systems. A stream of solids always behaves as a macrofluid, but for gas reacting with liquid, either phase can be a macro- or microfluid depending on the contacting scheme being used. The sketches of Fig. show completely opposite behavior.

Earliness of Mixing

- The fluid elements of a single flowing stream can mix with each other either early or late in their flow through the vessel.



- Usually this factor has little effect on overall behavior for a single flowing fluid.
- However, for a system with two entering reactant streams it can be very important.

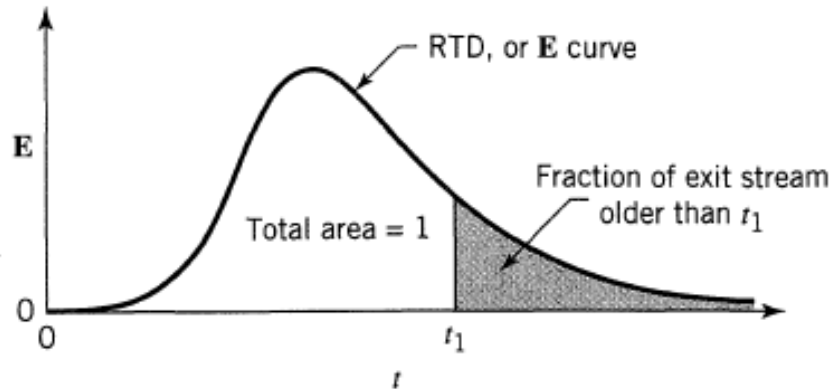


Role of RTD, State of Aggregation, and Earliness of Mixing in Determining Reactor behavior

- In some situations one of these three factors can be ignored; in others it can become crucial.
- Often, much depends on the time for reaction, t_{rx} the time mixing \bar{t}_{mix} , and the time for stay in the vessel \bar{t}_{stay} . In many cases \bar{t}_{stay} has a meaning somewhat like \bar{t}_{mix} but somewhat broader.

E, THE AGE DISTRIBUTION OF FLUID, THE RTD

- The distribution of these times for the stream of fluid leaving the vessel is called the exit age distribution E, or the residence time distribution RTD of fluid.
- E has the units of time^{-1} .
- We find it convenient to represent the RTD in such a way that the area under the curve is unity,



$$\int_0^{\infty} \mathbf{E} dt = 1$$

- This procedure is called normalizing the distribution.
- We should note one restriction on the E curve-that the fluid only enters and only leaves the vessel one time.
- This means that there should be no flow or diffusion or up flow eddies at the entrance or at the vessel exit.
- We call this the **closed vessel** boundary condition.
- Where elements of fluid can cross the vessel boundary more than one time we call this the **open vessel** boundary condition.
- With this representation the fraction of exit stream of age between t and $t + dt$ is

- the fraction younger than age t_1 is $\int_0^{t_1} \mathbf{E} dt$

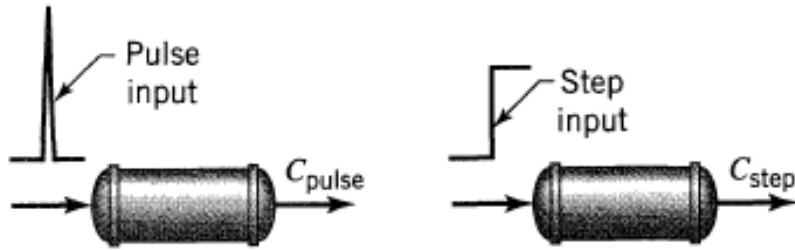
whereas the fraction of material older than t_1 , shown as the shaded area in

$$\int_{t_1}^{\infty} \mathbf{E} dt = 1 - \int_0^{t_1} \mathbf{E} dt$$

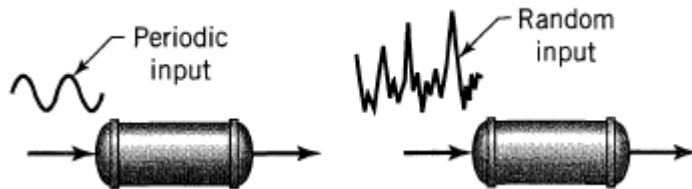
The \mathbf{E} curve is the distribution needed to account for nonideal flow.

Experimental Methods (Nonchemical) for Finding \mathbf{E}

- The simplest and most direct way of finding the \mathbf{E} curve uses a physical or nonreactive tracer.
- For special purposes, however, we may want to use a reactive tracer. This chapter deals in detail with the nonreactive tracer, and for this all sorts of experiments can be used.
- the pulse and the step experiments are easier to interpret,
- the periodic and random harder,
 here we only consider the pulse and the step experiment.
- We then show how to find reactor behavior knowing the \mathbf{E} curve for the reactor.



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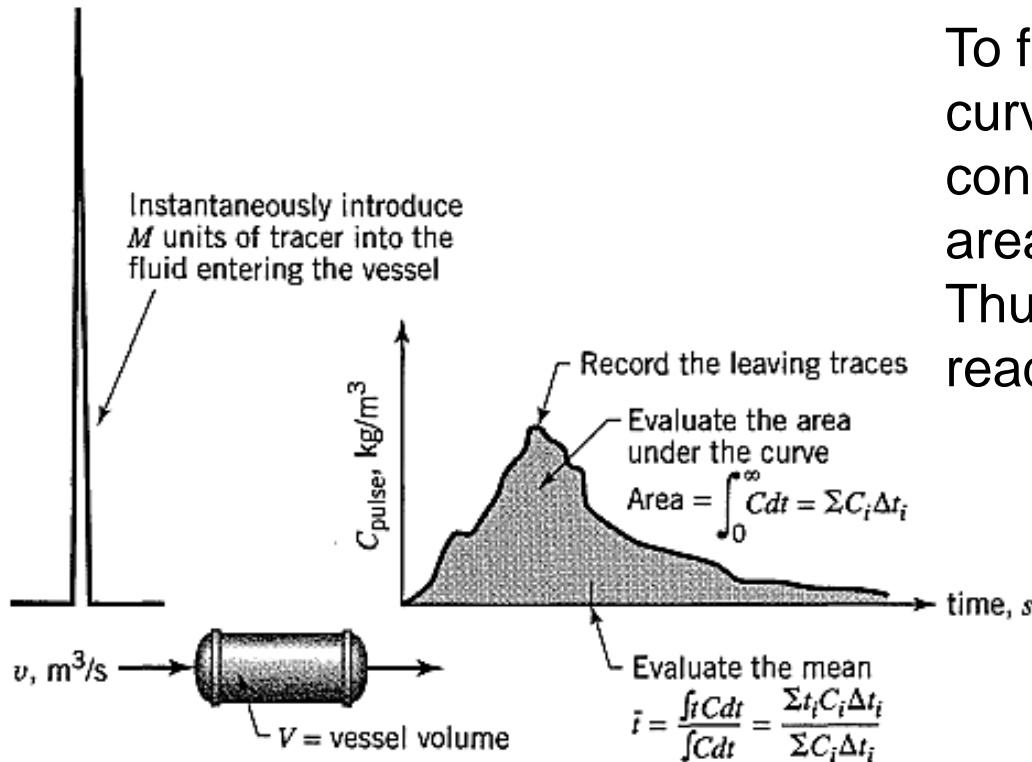


the periodic and random harder

The Pulse Experiment (single time fixed amount inlet)

- Let us find the E curve for a vessel of volume $V \text{ m}^3$ through which flows $v \text{ m}^3/\text{s}$ of fluid.
- For this instantaneously introduce M units of tracer (kg or moles) into the fluid entering the vessel,
- Record the concentration-time of tracer leaving the vessel.

- This is the C_{pulse} curve.
- From the material balance for the vessel we find



To find the E curve from the C_{pulse} curve simply change the concentration scale such that the area under the curve is unity. Thus, simply divide the concentration readings by M/v ,

$$\mathbf{E} = \frac{C_{\text{pulse}}}{M/v}$$

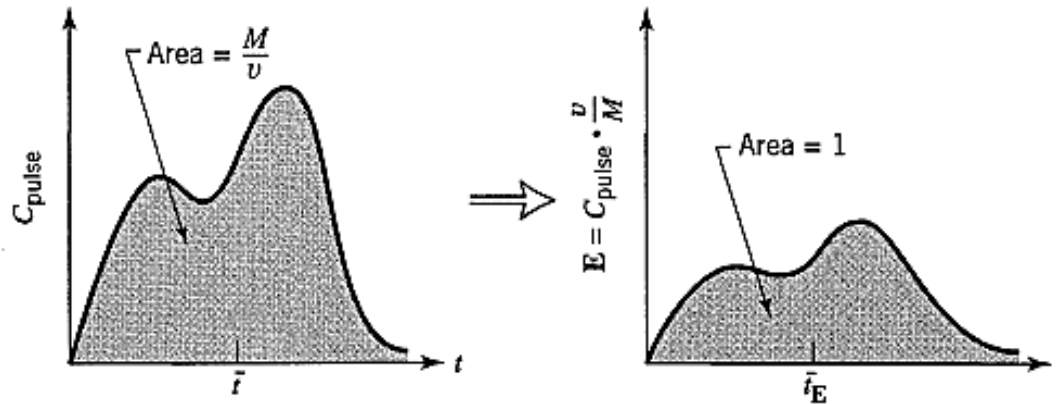
(Area under the C_{pulse} curve):

$$A = \int_0^{\infty} C dt \cong \sum_i C_i \Delta t_i = \frac{M}{v} \quad \left[\frac{\text{kg} \cdot \text{s}}{\text{m}^3} \right]$$

(Mean of the C_{pulse} curve):

$$\bar{t} = \frac{\int_0^{\infty} tC dt}{\int_0^{\infty} C dt} \cong \frac{\sum_i t_i C_i \Delta t_i}{\sum_i C_i \Delta t_i} = \frac{V}{v} \quad [\text{s}]$$

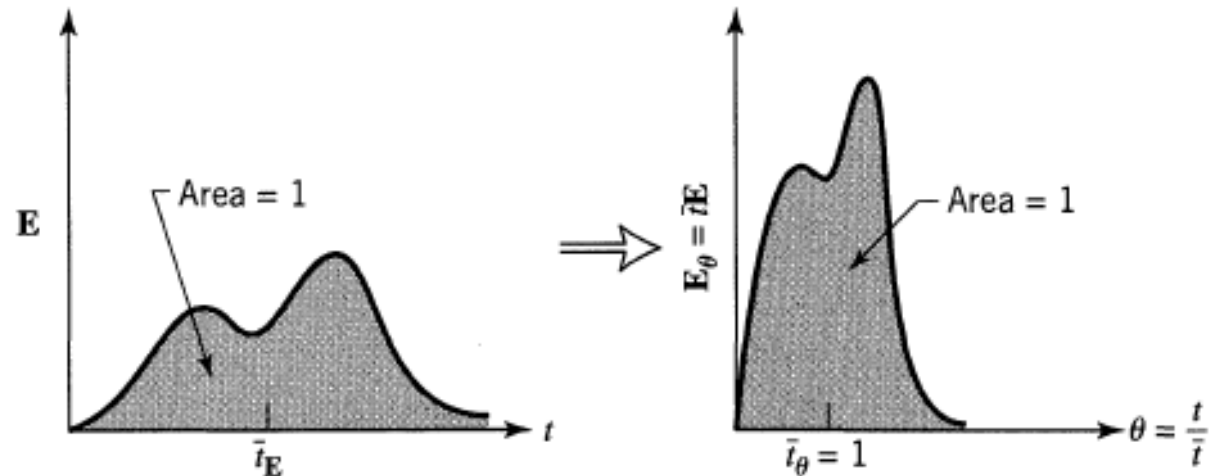
Transforming an experimental Cpulse curve into an E curve



We have another RTD function E_θ . Here time is measured in terms of mean residence time $\theta = t/\bar{t}$. Thus

Transforming an E curve into an E_θ curve.

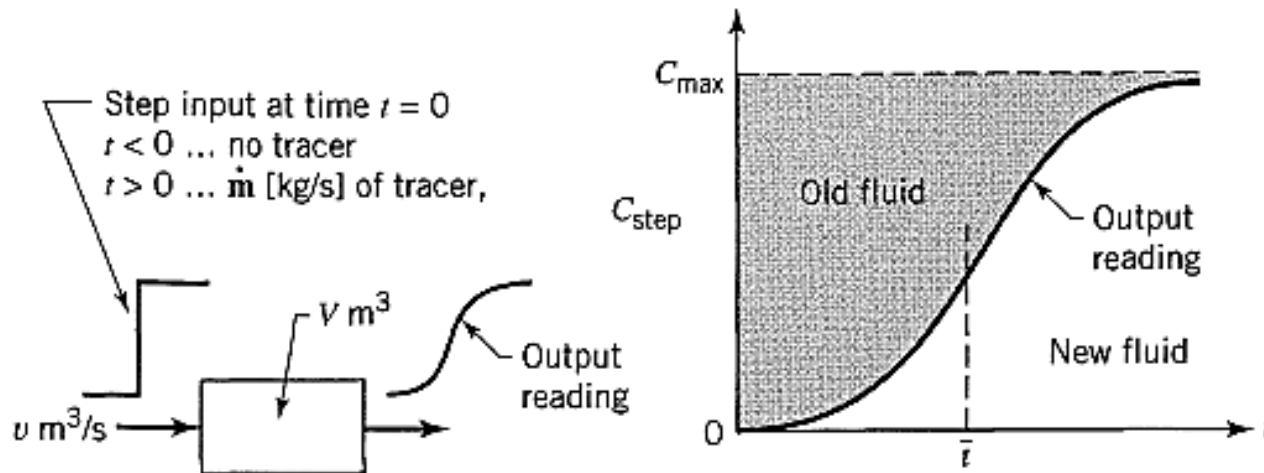
$$E_\theta = \bar{t}E = \frac{V}{v} \cdot \frac{C_{\text{pulse}}}{M/v} = \frac{V}{M} C_{\text{pulse}}$$



The Step Experiment

Consider $v \text{ m}^3/\text{s}$ of fluid flowing through a vessel of volume V . Now at time $t = 0$ switch from ordinary fluid to fluid with tracer of concentration $C_{\text{max}} = \left[\frac{\text{kg or mol}}{\text{m}^3} \right]$, and measure the outlet tracer concentration C_{step} versus t , as shown

Information obtainable from a step tracer experiment.



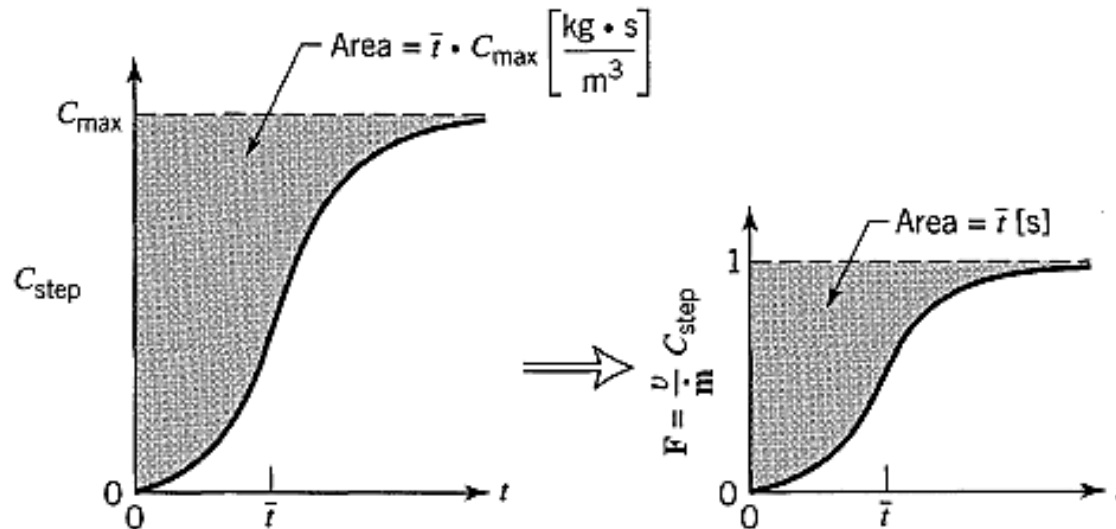
A material balance relates the different measured quantities of the output curve of a step input

$$\text{shaded area} = C_{\text{max}} \bar{t} = \frac{\dot{m} V}{v^2} \left[\frac{\text{kg} \cdot \text{s}^2}{\text{m}^3} \right]$$

$$C_{\text{max}} = \frac{\dot{m}}{v} \left[\frac{\text{kg} \cdot \text{s}}{\text{m}^3} \right]$$

$$\bar{t} = \frac{\int_0^{C_{\text{max}}} t dC_{\text{step}}}{\int_0^{C_{\text{max}}} dC_{\text{step}}} = \frac{1}{C_{\text{max}}} \int_0^{C_{\text{max}}} t dC_{\text{step}}$$

- Transforming an experimental C_{step} curve to an F curve
- The dimensionless form of the C_{step} curve is called the **F** curve.
- It is found by having the tracer concentration rise from zero to unity, as shown in Fig



Relationship between the F and E Curves

To relate E with **F** imagine a steady flow of white fluid.

Then at time $t = 0$ switch to red and record the rising concentration of red fluid in the exit stream, the F curve

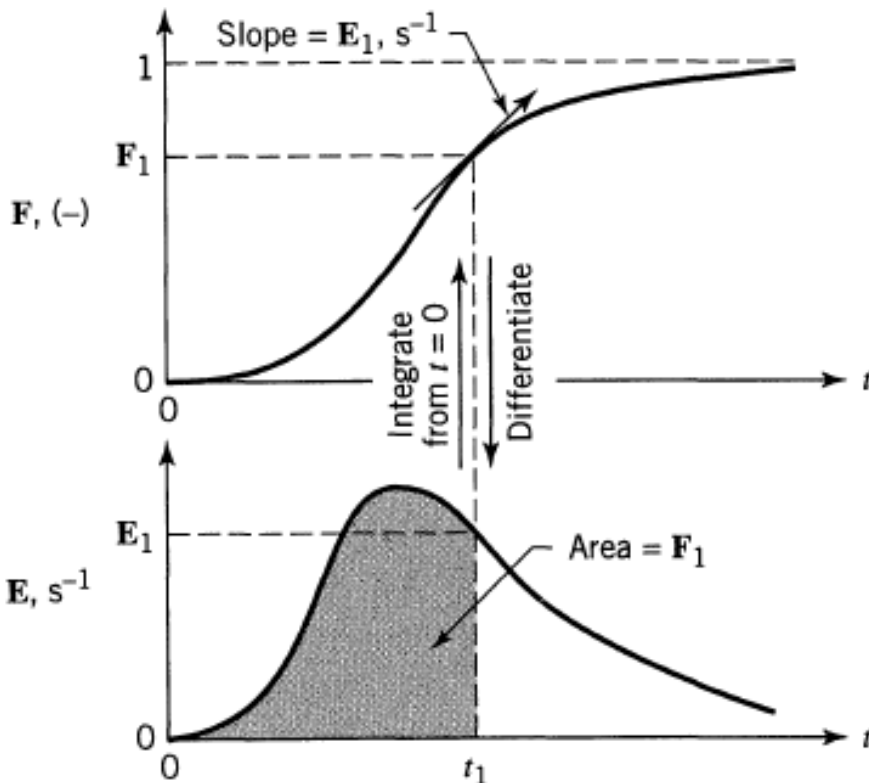
- At any time $t > 0$ red fluid and only red fluid in the exit stream is younger than age t . Thus we have

$$\left(\begin{array}{c} \text{fraction of red fluid} \\ \text{in the exit stream} \end{array} \right) = \left(\begin{array}{c} \text{fraction of exit stream} \\ \text{younger than age } t \end{array} \right)$$

$$\mathbf{F} = \int_0^t \mathbf{E} dt$$

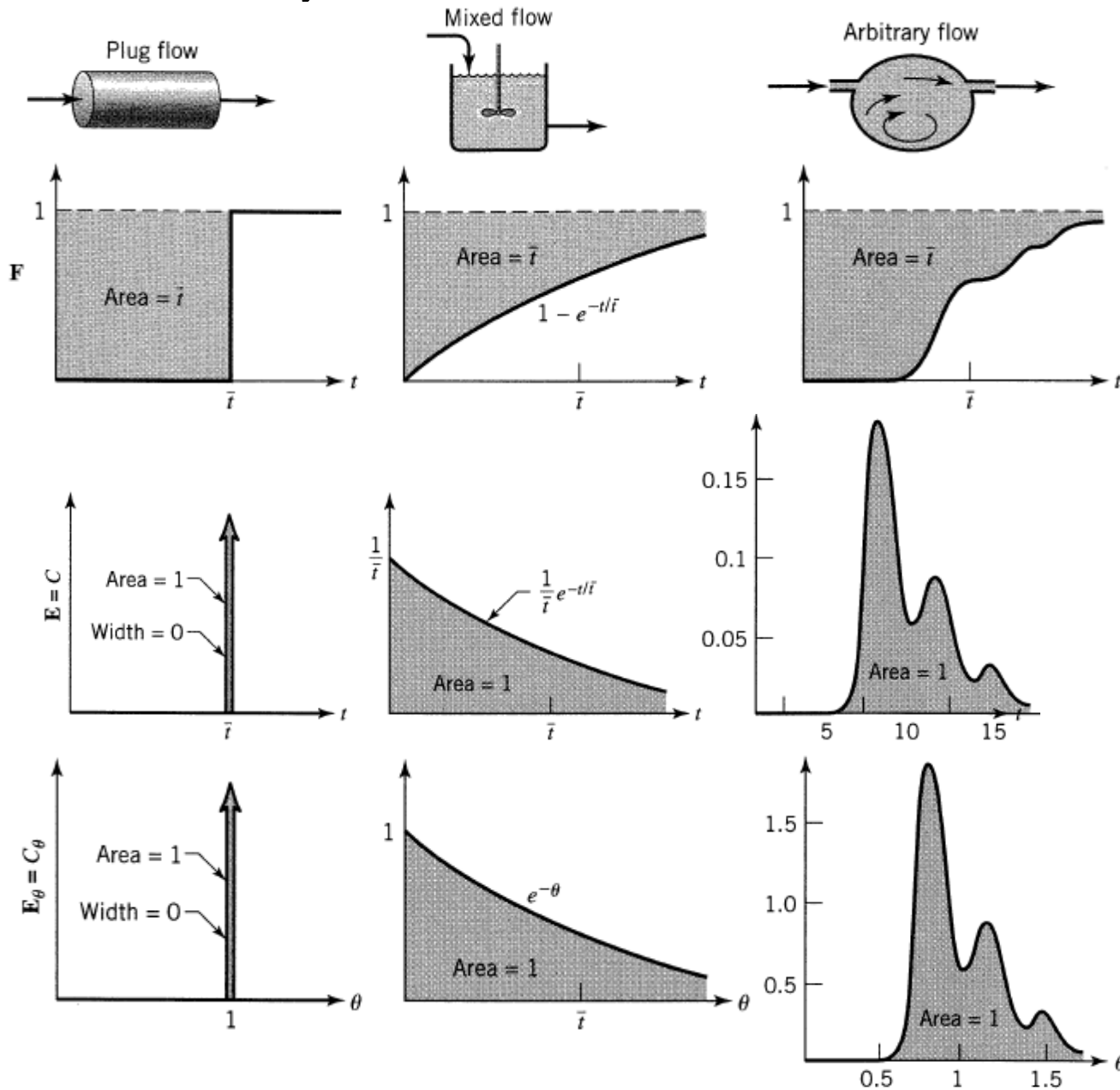
and on differentiating

$$\frac{d\mathbf{F}}{dt} = \mathbf{E}$$



- These relationships show how stimulus-response experiments, using either step or pulse inputs can conveniently give the RTD and mean flow rate of fluid in the vessel.
- We should remember that these relationships only hold for closed vessels.
- When this boundary condition is not met, then the C_{puls} and E curves differ.

- Properties of the E and F curves for various flows. Curves are drawn in terms of ordinary and dimensionless time units.



At any time these curves are related as follows:

$$\mathbf{E} = \frac{v}{\dot{\mathbf{m}}} \cdot C_{\text{pulse}}, \quad \mathbf{F} = \frac{v}{\dot{\mathbf{m}}} \cdot C_{\text{step}}, \quad \mathbf{E} = \frac{d\mathbf{F}}{dt},$$

$$\bar{t} = \frac{V}{v}, \quad \theta = \frac{t}{\bar{t}}, \quad \bar{\theta}_{\mathbf{E}} = 1, \quad \mathbf{E}_{\theta} = \bar{t}\mathbf{E}$$

θ , \mathbf{E}_{θ} , \mathbf{F} ... all dimensionless, $\mathbf{E} = [\text{time}^{-1}]$

The concentration readings in Table E11.1 represent a continuous response to a pulse input into a closed vessel which is to be used as a chemical reactor. Calculate the mean residence time of fluid in the vessel t , and tabulate and plot the exit age distribution \mathbf{E} .

Table E11.1

Time t , min	Tracer Output Concentration, C_{pulse} gm/liter fluid
0	0
5	3
10	5
15	5
20	4
25	2
30	1
35	0

$$\bar{t} = \frac{\sum t_i C_i \Delta t_i}{\sum C_i \Delta t_i} \quad \frac{\Delta t = \text{constant}}{\sum C_i \Delta t_i} = \frac{\sum t_i C_i}{\sum C_i}$$

$$\text{Area} = \sum C \Delta t :$$

$$\mathbf{E} = \frac{C}{\text{area}}$$

The mean residence time, from Eq. 4, is

$$\bar{t} = \frac{\sum t_i C_i \Delta t_i}{\sum C_i \Delta t_i} \quad \Delta t = \text{constant} \quad \frac{\sum t_i C_i}{\sum C_i}$$

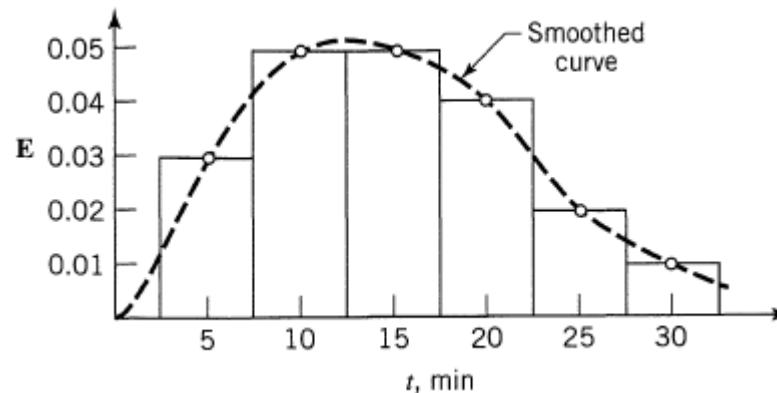
$$= \frac{5 \times 3 + 10 \times 5 + 15 \times 5 + 20 \times 4 + 25 \times 2 + 30 \times 1}{3 + 5 + 5 + 4 + 2 + 1} = 15 \text{ min}$$

The area under the concentration-time curve,

$$\text{Area} = \sum C \Delta t = (3 + 5 + 5 + 4 + 2 + 1)5 = 100 \text{ gm} \cdot \text{min/liter}$$

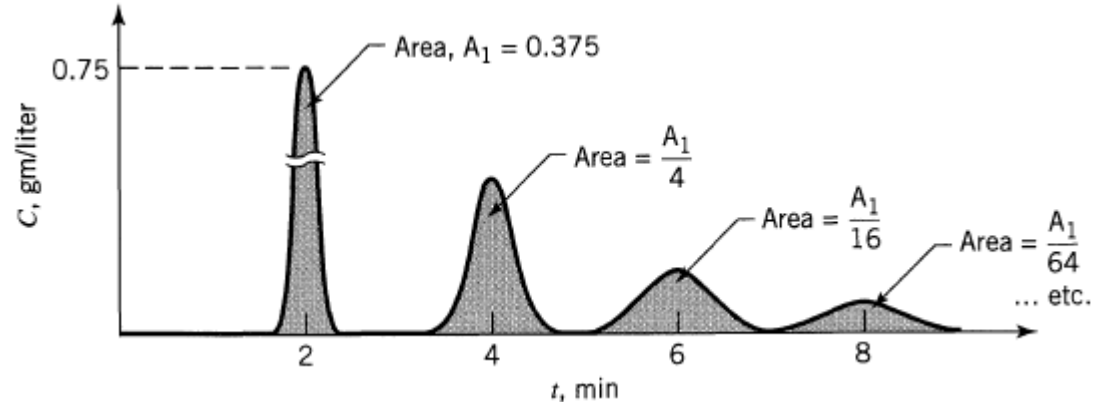
gives the total amount of tracer introduced. To find **E**, the area under this curve must be unity; hence, the concentration readings must each be divided by the total area, giving

$\mathbf{E} = \frac{C}{\text{area}}, \text{min}^{-1}$	t, min	0	5	10	15	20	25	30
		0	0.03	0.05	0.05	0.04	0.02	0.01



A large tank (860 liters) is used as a gas-liquid contactor. Gas bubbles up through the vessel and out the top, liquid flows in at one part and out the other at 5 liters/s. To get an idea of the flow pattern of liquid in this tank a pulse of tracer ($M = 150$ gm) is injected at the liquid inlet and measured at the outlet, as shown

- Is this a properly done experiment?
- If so, find the liquid fraction in the vessel.
- Determine the E curve for the liquid.
- Qualitatively what do you think is happening in the vessel?



- Check the material balance against the tracer curve. From the material balance, Eq. 3, we should have

$$\text{Area} = \frac{M}{v} = \frac{150 \text{ gm}}{5 \text{ liters/s}} = 30 \frac{\text{gm} \cdot \text{s}}{\text{liter}} = 0.5 \frac{\text{gm} \cdot \text{min}}{\text{liter}}$$

$$\text{Area} = A_1 \left(1 + \frac{1}{4} + \frac{1}{16} + \dots \right) = 0.375 \left(\frac{4}{3} \right) = 0.5 \frac{\text{gm} \cdot \text{min}}{\text{liter}}$$

$$\bar{t}_l = \frac{\int tC dt}{\int C dt} = \frac{1}{0.5} \left[2A_1 + 4 \times \frac{A_1}{4} + 6 \times \frac{A_1}{16} + 8 \times \frac{A_1}{64} + \dots \right] = 2.67 \text{ min}$$

Thus the liquid volume in the vessel is

$$V_l = \bar{t}_l v_l = 2.67(5 \times 60) = 800 \text{ liters}$$

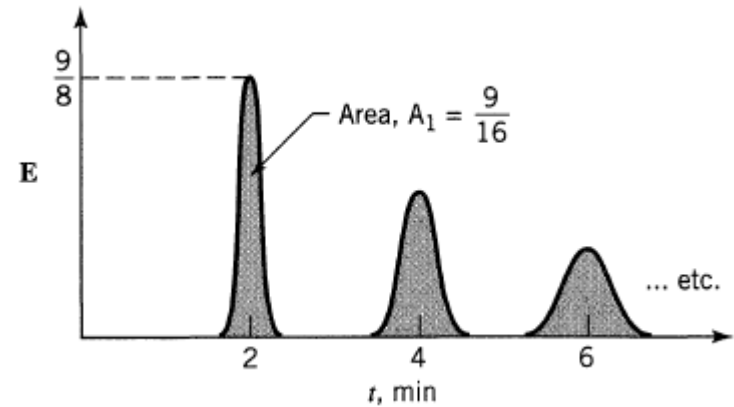
and the volume fraction of phases is

$$\left. \begin{aligned} \text{Fraction of liquid} &= \frac{800}{860} = 93\% \\ \text{Fraction of gas} &= 7\% \end{aligned} \right\}$$

(c) Finally, from Eq. 5 we find the **E** curve, or

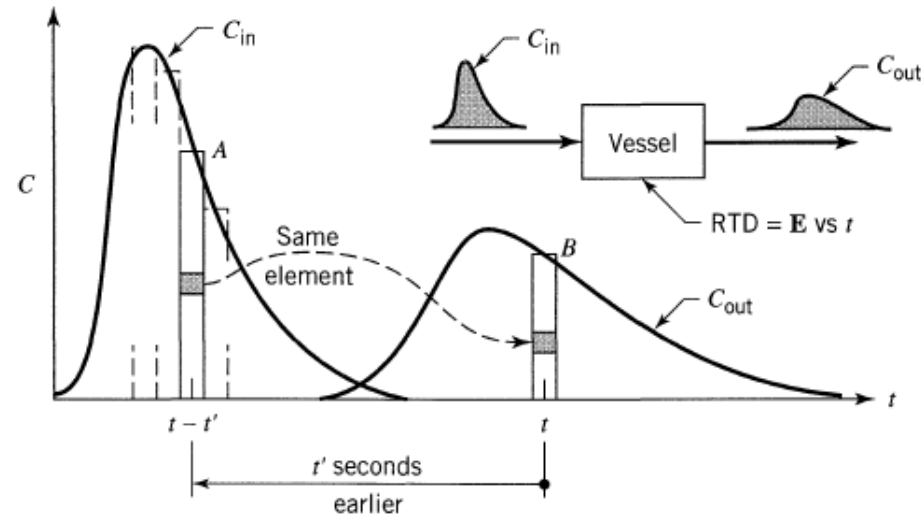
$$\mathbf{E} = \frac{C_{\text{pulse}}}{M/v} = \frac{0.75}{0.5} C = 1.5 C$$

Thus the **E** curve for the liquid is as shown in Fig.



The Convolution Integral

- Suppose we introduce into a vessel a one-shot tracer signal C_{in} versus t as shown in Fig.



- In passing through the vessel the signal will be modified to give an output signal C_{out} versus t .
- Since the flow with its particular RTD is responsible for this modification let us relate C_{in} , E , and C_{out} .
- Focus attention on tracer leaving at time about t . This is shown as the narrow rectangle B in Fig. 11.15. We may then write

$$\left(\begin{array}{l} \text{tracer leaving} \\ \text{in rectangle } B \end{array} \right) = \left(\begin{array}{l} \text{all the tracer entering } t' \text{ seconds earlier than } t, \\ \text{and staying for time } t' \text{ in the vessel} \end{array} \right)$$

the tracer which enters t' seconds earlier than t as the narrow rectangle A. In terms of this rectangle the above equation may be written

$$\left(\begin{array}{l} \text{tracer leaving} \\ \text{in rectangle } B \end{array} \right) = \sum_{\substack{\text{all rectangles} \\ A \text{ which enter} \\ \text{earlier than} \\ \text{time } t}} \left(\begin{array}{l} \text{tracer in} \\ \text{rectangle} \\ A \end{array} \right) \left(\begin{array}{l} \text{fraction of tracer in } A \\ \text{which stays for about} \\ t' \text{ seconds in the vessel} \end{array} \right)$$

In symbols and taking limits (shrinking the rectangles) we obtain the desired relationship, which is called the convolution integral

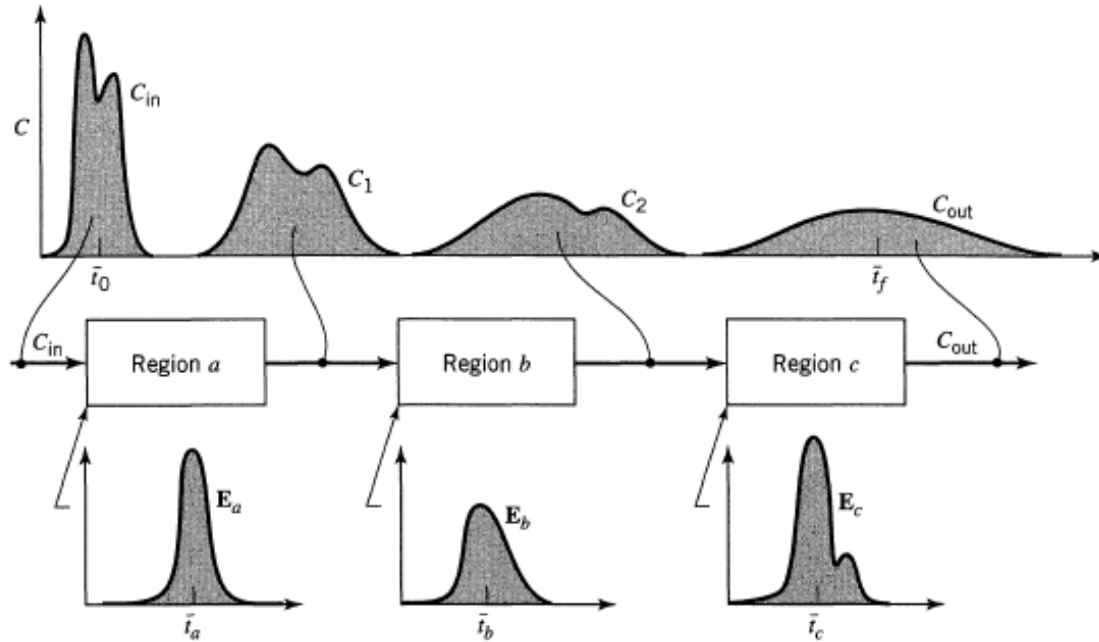
$$C_{\text{out}}(t) = \int_0^t C_{\text{in}}(t-t')\mathbf{E}(t')dt'$$

$$C_{\text{out}}(t) = \int_0^t C_{\text{in}}(t')\mathbf{E}(t-t')dt'$$

We say that C_{out} is the convolution of \mathbf{E} with C_{in} and we write concisely

$$C_{\text{out}} = \mathbf{E} * C_{\text{in}} \quad \text{or} \quad C_{\text{out}} = C_{\text{in}} * \mathbf{E}$$

Modification of an input tracer signal C_{in} on passing through three successive regions.



Application of These Tools.

To illustrate the uses of these mathematical tools consider three independent flow units a , b , and c , which are closed and connected in series

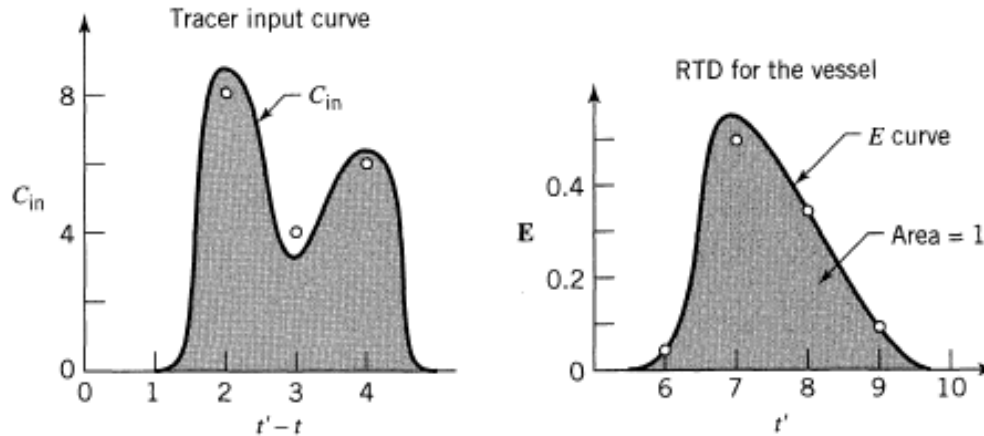
Problem 1. If the input signal C_{in} is measured and the exit age distribution functions E_a , E_b , and E_c are known, then C_1 is the convolution of E_a with C_{in} and so on, thus

$$C_1 = C_{in} * E_a, \quad C_2 = C_1 * E_b, \quad C_{out} = C_2 * E_c$$

$$C_{out} = C_{in} * E_a * E_b * E_c$$

Thus we can determine the output from a multiregion flow unit.

- Let us illustrate the use of the convolution equation, Eq. 10, with a very simple example in which we want to find C_{out} given C_{in} and the E curve for the vessel, as shown in Fig.



First of all, take 1 min time slices. The given data are then

$t' - t$	C_{in}	t'	E
0	0	5	0
1	0	6	0.05
2	8	7	0.50
3	4	8	0.35
4	6	9	0.10
5	0	10	0

Note: The area under the E curve is unity.

Now the first bit of tracer leaves at 8 min, the last bit at 13 min. Thus, applying the convolution integral, in discrete form, we have

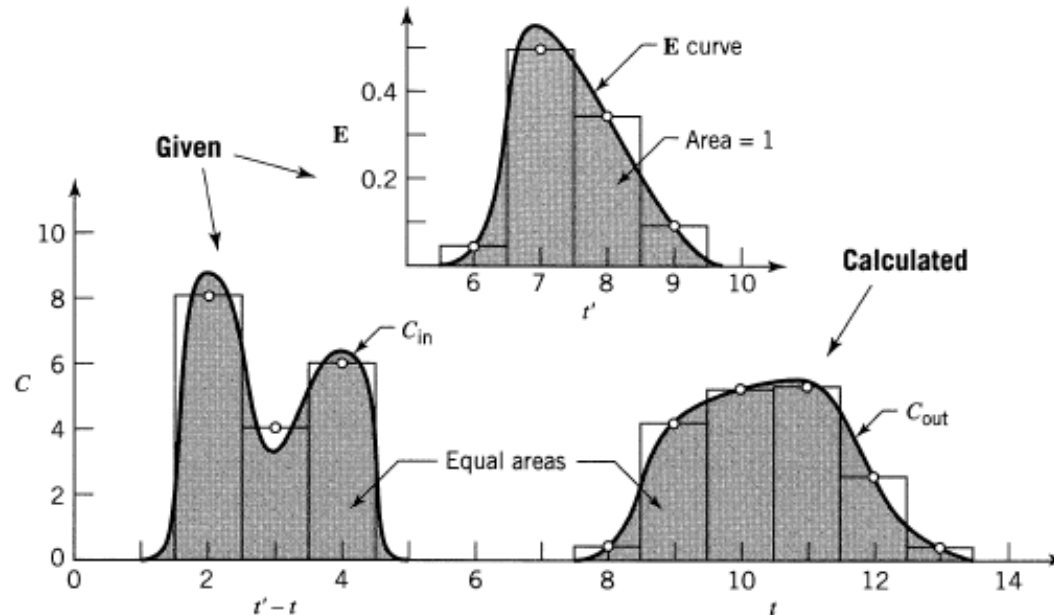
$$C_{out}(t) = \int_0^t C_{in}(t-t')E(t')dt'$$

$t' - t$	C_{in}
0	0
1	0
2	8
3	4
4	6
5	0

t'	E
5	0
6	0.05
7	0.50
8	0.35
9	0.10
10	0

t	C_{out}
7	0 = 0
8	$8 \times 0.05 = 0.4$
9	$8 \times 0.5 + 4 \times 0.05 = 4.2$
10	$8 \times 0.35 + 4 \times 0.5 + 6 \times 0.05 = 5.1$
11	$8 \times 0.10 + 4 \times 0.35 + 6 \times 0.5 = 5.2$
12	$4 \times 0.10 + 6 \times 0.35 = 2.5$
13	$6 \times 0.10 = 0.6$
14	0 = 0

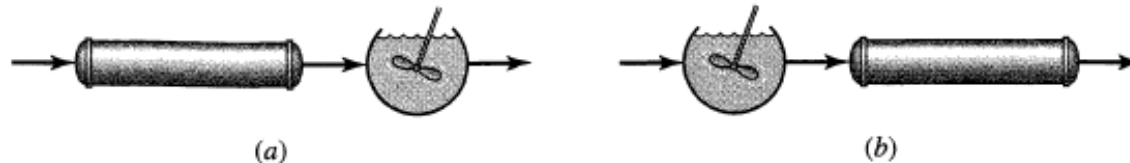
The C_{in} , E , and C_{out} curves, in discrete and in continuous form, are shown in Fig. Note that the area under the C_{out} curve equals the area under the C_{in} curve.



CONVERSION IN NON-IDEAL FLOW REACTORS

- To evaluate reactor behavior in general we have to know four factors:
 1. the kinetics of the reaction
 2. the RTD of fluid in the reactor
 3. the earliness or lateness of fluid mixing in the reactor
 4. whether the fluid is a micro or macro fluid

To consider the early and late mixing of a microfluid, consider the two flow patterns shown in Fig. for a reactor processing a second-order reaction.



(a) the reactant starts at high concentration and reacts away rapidly because $n > 1$.

(b) the fluid drops immediately to a low concentration.

Since the rate of reaction drops more rapidly than does the concentration you will end up with a lower conversion.

Thus, for microfluids Late mixing favors reactions where $n > 1$

Early mixing favors reactions where $n < 1$

For macrofluids, imagine little clumps of fluid staying for different lengths of time in the reactor (given by the E function). Each clump reacts away as a little batch reactor, thus fluid elements will have different compositions.

So the mean composition in the exit stream will have to account for these two factors, the kinetics and the RTD. In words, then

$$\left(\begin{array}{c} \text{mean concentration} \\ \text{of reactant} \\ \text{in exit stream} \end{array} \right) = \sum_{\text{all elements of exit stream}} \left(\begin{array}{c} \text{concentration of} \\ \text{reactant remaining} \\ \text{in an element of} \\ \text{age between } t \\ \text{and } t + dt \end{array} \right) \left(\begin{array}{c} \text{fraction of exit} \\ \text{stream which is} \\ \text{of age between } t \\ \text{and } t + dt \end{array} \right)$$

$$\left(\frac{\bar{C}_A}{C_{A0}} \right)_{\text{at exit}} = \int_0^{\infty} \left(\frac{C_A}{C_{A0}} \right)_{\text{for an element or little batch of fluid of age } t} \cdot \mathbf{E} dt$$

or in terms of conversions

$$\bar{X}_A = \int_0^{\infty} (X_A)_{\text{element}} \cdot \mathbf{E} dt$$

or in a form suitable for numerical integration

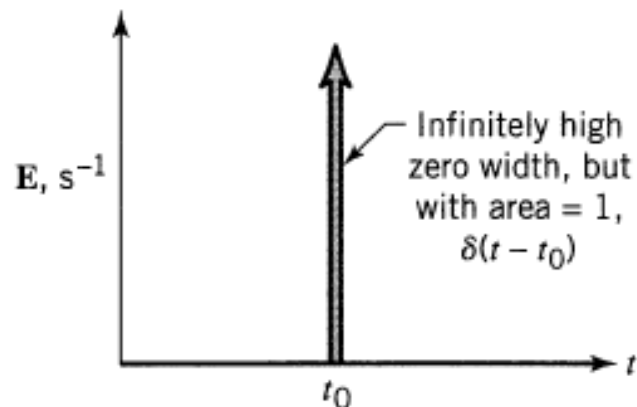
$$\frac{\bar{C}_A}{C_{A0}} = \sum_{\text{all age intervals}} \left(\frac{C_A}{C_{A0}} \right)_{\text{element}} \cdot \mathbf{E} \Delta t$$

- for first-order reactions $\left(\frac{C_A}{C_{A0}}\right)_{\text{element}} = e^{-kt}$
- for second-order reactions $\left(\frac{C_A}{C_{A0}}\right)_{\text{element}} = \frac{1}{1 + kC_{A0}t}$
- for an n th-order reaction $\left(\frac{C_A}{C_{A0}}\right)_{\text{element}} = [1 + (n - 1)C_{A0}^{n-1}kt]^{1/1-n}$

The Dirac Delta Function, $\delta(t - t_0)$. One **E** curve which may puzzle us is the one which represents plug flow. We call this the Dirac δ function, and in symbols we show it as

$$\delta(t - t_0)$$

which says that the pulse occurs at $t = t_0$, as seen in Fig. The two properties of this function which we need to know are



Area under the curve: $\int_0^{\infty} \delta(t - t_0) dt = 1$

Any integration with a δ function: $\int_0^{\infty} \delta(t - t_0) f(t) dt = f(t_0)$

Once we understand what this means we will see that it is easier to integrate with a δ function than with any other. For example,

$$\int_0^{\infty} \delta(t - 5)t^6 dt = 5^6 \quad (\text{just replace } t_0 \text{ by } 5)$$

$$\int_0^3 \delta(t - 5)t^6 dt = 0$$

CONVERSION IN REACTORS HAVING NON-IDEAL FLOW

Find the fraction of reactant unconverted in the real reactor and compare this with the fraction unconverted in a plug flow reactor of the same size.

$$-r_A = kC_A, \quad k = 0.307 \text{ min}^{-1}$$

For the plug **flow** reactor with negligible density change we have and with

$$\tau = C_{A0} \int_0^{X_A} \frac{dX_A}{-r_A} = -\frac{1}{k} \int_{C_{A0}}^{C_A} \frac{dC_A}{C_A} = \frac{1}{k} \ln \frac{C_{A0}}{C_A}$$

$$\frac{C_A}{C_{A0}} = e^{-k\tau} = e^{-(0.307)(15)} = e^{-4.6} = \underline{\underline{0.01}}$$

For real system

$$\left(\frac{\bar{C}_A}{C_{A0}}\right)_{\text{at exit}} = \int_0^{\infty} \left(\frac{C_A}{C_{A0}}\right)_{\text{for an element or little batch of fluid of age } t} \cdot \mathbf{E} dt$$

or in terms of conversions

$$\bar{X}_A = \int_0^{\infty} (X_A)_{\text{element}} \cdot \mathbf{E} dt$$

or in a form suitable for numerical integration

$$\frac{\bar{C}_A}{C_{A0}} = \sum_{\substack{\text{all age} \\ \text{intervals}}} \left(\frac{C_A}{C_{A0}}\right)_{\text{element}} \cdot \mathbf{E} \Delta t$$

For the *real reactor* the fraction unconverted, given by above Eq. for macrofluids, is found in Table. Hence the fraction of reactant unconverted in the real reactor

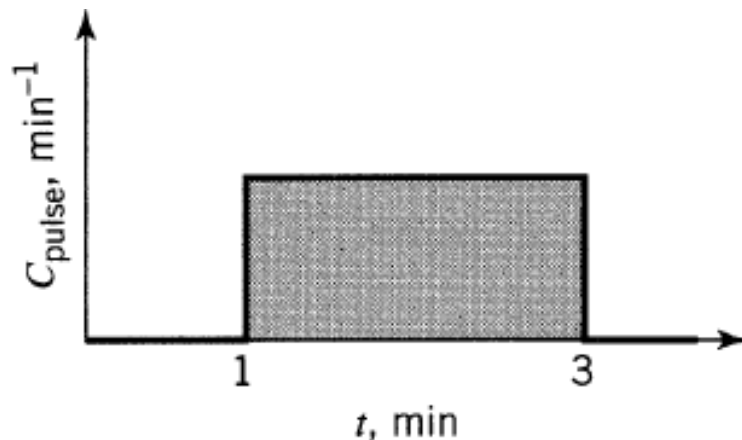
t	\mathbf{E}	kt	e^{-kt}	$e^{-kt}\mathbf{E} \Delta t$
5	0.03	1.53	0.2154	(0.2154)(0.03)(5) = 0.0323
10	0.05	3.07	0.0464	0.0116
15	0.05	4.60	0.0100	0.0025
20	0.04	6.14	0.0021	0.0004
25	0.02	7.68	0.0005	0.0001
30	0.01	9.21	0.0001	0
$\underbrace{\hspace{10em}}_{\text{given}}$				$\frac{C_A}{C_{A0}} = \sum e^{-kt}\mathbf{E} \Delta t = \underline{\underline{0.0469}}$

- Note that since this is a first-order reaction we can treat it as a microfluid, or a macrofluid, whatever we wish.
- In this problem we solved the plug flow case as a microfluid, and we solved the non-ideal case as a macrofluid.

REACTION OF A MACROFLUID

Dispersed noncoalescing droplets ($C_{A0} = 2$ mol/liter) react ($A \rightarrow R$, $-r_A = kC_A^2$, $k = 0.5$ liter/mol \cdot min) as they pass through a contactor. Find the average concentration of A remaining in the droplets leaving the contactor if their RTD is given by the curve in Fig

is given by the curve in Fig



Equation is the pertinent performance equation. Evaluate terms in this expression. For

$$-r_A = kC_A^2, \quad k = 0.5 \text{ liter/mol} \cdot \text{min}$$

$$\frac{C_A}{C_{A0}} = \frac{1}{1 + kC_{A0}t} = \frac{1}{1 + 0.5(2)t} = \frac{1}{1 + t}$$

$$E = C_{A0}/\text{Area}$$

$$\text{Area} = 2 \times 2 = 4$$

$$E = 2/4 = 0.5$$

With $E = 0.5$ for $1 < t < 3$,

$$\frac{\bar{C}_A}{C_{A0}} = \int_0^\infty \left(\frac{C_A}{C_{A0}} \right)_{\text{batch}} \mathbf{E} dt = \int_1^3 \frac{1}{1+t} \cdot (0.5) dt = 0.5 \ln 2 = 0.347$$

$$\bar{X}_A = 1 - 0.347 = 0.653, \quad \text{or} \quad \underline{\underline{65\%}}$$