Convergence of a Series of Well-Stirred Reactors to Plug-Flow Reactor

Paul Molin and Patrick Gervais
Laboratoire Génie des Procédés Alimentaires et Biotechnologiques, ENS.BANA, 21000 Dijon, France

In industrial processes, the characterization of the reactors used is very important for many reasons such as quality and homogenity of the products resulting from the process, speed of the process, and maintenance of the reactors (Moser, 1981). The study of the residence time distribution (RTD) allows the understanding of the distributions of the solutes (Gervais et al., 1986). Among all the different types of reactors, the two extreme behaviors are those of the well-stirred reactor (CSTR) and the plug-flow reactor (PFR). The convergence of a series of well-stirred reactors to a plug-flow reactor is intuitive, however a mathematical proof is more difficult since this convergence is not uniform. Such a demonstration was recently made by using the Laplace transform, which proves the convergence in the Laplace universe (Villermaux, 1993). Nevertheless in such a case, the convergence of the series of inverse Laplace transforms is not trivial and has not yet been proven. We show here directly the convergence of the series in the time variable.

RTD for the Plug-Flow and the Well-Stirred Reactors

The characterization of a reactor can be carried out by introducing at $t = 0$ a tracer at an initial concentration C_0 which corresponds to a step change input. The measurement of the outlet concentration allows the determination of the RTD (Villermaux, 1993).

In the case of a plug-flow reactor, all the particles of the tracer having the same residence time, the concentration on exit takes also the form of a unit step, shifted in time. The shift in time is equal to:

$$\tau = \frac{V}{Q}$$

where V is the volume of the reactor, and Q is the flow rate of the solution through the reactor.

The case of a well-stirred reactor is described by the mass balance of the tracer during a little time interval dt, when no chemical reaction or transformation of the solute occurs in the reactor. This mass balance results into the differential equation:

$$\tau \frac{dC}{dt} + C = C_0$$

which has the solution, with initial condition $C = 0$:

$$\frac{C}{C_0} = 1 - e^{-\frac{t}{\tau}},$$

where C is the internal (and outlet) solute concentration.

Case of a Series of Well-Stirred Reactors

Figure 1 shows a series of well-stirred reactors and some responses to a unit step, depending on the number of reactors. Each reactor has a volume equal to V/n, where V is the global volume of the solution in the process, and n is the number of reactors.

The equation for the ith reactor is:

$$\frac{\tau}{n} \frac{dC_i}{dt} + C_i = C_{i-1} \quad i = 1 \ldots n$$

Where C_i is the solute concentration in the ith reactor; $\tau/n = V/nQ$ is the time constant of the ith reactor.

The resolution of this system of linear equations gives the outlet concentration of the solution in the last tank (Figure 1) (Loncin, 1976):

$$C_n = C_0 \left[1 - e^{-\frac{nt}{\tau}} \left\{ 1 + n\frac{t}{\tau} + \frac{1}{2!}\left(\frac{nt}{\tau}\right)^2 + \cdots + \frac{1}{(n\text{-}1)!}\left(\frac{nt}{\tau}\right)^{n-1} \right\} \right].$$

Correspondence concerning this article should be addressed to P. Molin.

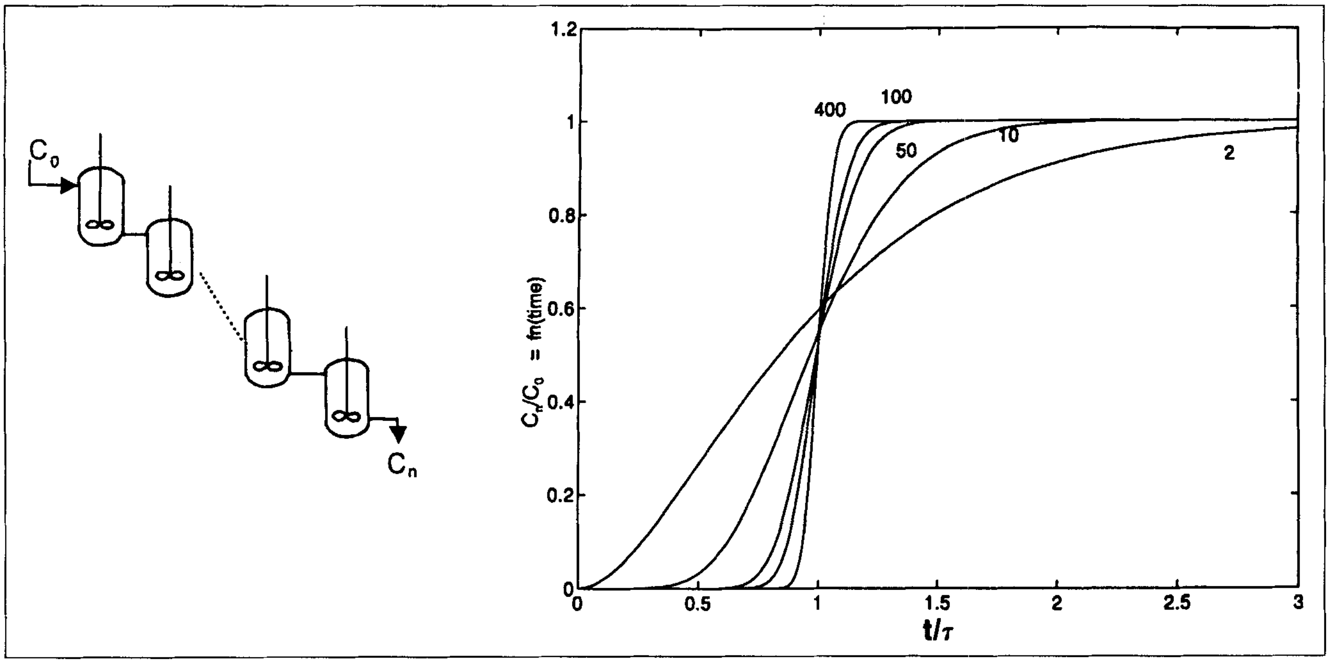

Figure 1. Series of continuous stirred tank reactors and their responses to a step input of concentration depending on the number of reactors.

Problem of Convergence

We will prove that the function $C_n/C_0 = f_n(t)$ tends towards a unit step when n tends towards infinity, the change point of this unit step being the time τ.

According to the first paragraph, this will exactly be the description of the response of a plug-flow reactor to a unit step input.

Transformation of $f_n(t)$

The Taylor formula with integral remainder for the exponential function calculated at the point nt/τ gives:

$$e^{\frac{nt}{\tau}} = \sum_{j=0}^{n-1} \frac{1}{j!}\left(\frac{nt}{\tau}\right)^j + \frac{1}{(n-1)!}\int_0^{\frac{nt}{\tau}} \left(\frac{nt}{\tau} - x\right)^{n-1} e^x\, dx.$$

Then

$$f_n(t) = \frac{e^{-\frac{nt}{\tau}}}{(n-1)!}\int_0^{\frac{nt}{\tau}} \left(\frac{nt}{\tau} - x\right)^{n-1} e^x\, dx = \frac{(n\alpha)^n}{n!} e^{-n\alpha} + \frac{1}{n!}\int_0^{n\alpha} u^n e^{-u}\, du$$

(by the changes of $u = nt/\tau - x$ and $\alpha = t/\tau$).

By the use of the Stirling formula, the first term is equivalent to

$$\frac{(n\alpha)^n}{n!} e^{-n\alpha} \sim \frac{n^n\alpha^n e^{-n\alpha}}{n^n e^{-n}\sqrt{2\pi n}} = \frac{e^{-n(\alpha - 1 - \log\,\alpha)}}{\sqrt{2\pi n}}$$

The function $\alpha - 1 - \log\,\alpha$ being positive or zero for all values of α, this last term tends towards 0 uniformly for all values of α.

Let us now examine the second term, called $I_n(\alpha)$:

$$I_n(\alpha) = \frac{1}{n!}\int_0^{n\alpha} u^n e^{-u}\, du.$$

Introducing $v = u/n\alpha$, we obtain:

$$I_n(\alpha) = \frac{1}{n!}\int_0^1 (n\alpha v)^n e^{-n\alpha v} n\alpha\, dv = \frac{n^{n+1}\alpha^{n+1}}{n!}\int_0^1 v^n e^{-n\alpha v}\, dv.$$

Let:

$$J_n(\alpha) = \int_0^1 v^n e^{-n\alpha v} dv = \int_0^1 e^{n(\log v - \alpha v)} dv = \int_0^1 e^{nh(v)}\, dv.$$

The function $h(v)$ has a second derivative $h'' = -1/v^2$ always negative, it has a maximum at the point $1/\alpha$, with a value at this point equal to $-1 - \log\alpha$, and it tends towards $-\infty$ when v tends towards 0 or $+\infty$.

Convergence

We will discuss the convergence according to the value of α with respect to 1, which corresponds to the value of t with respect to τ.

$\alpha < 1$. In this case, the maximum of $h(v)$ lies beyond the upper value of the integration interval, and $h(v) \le h(1)$, that is, $h(v) \leq -\alpha$ for all $v \in [0, 1]$. Then we obtain the following inequalities:

$$J_n(\alpha) \leq e^{-n\alpha} \quad \text{and} \quad I_n(\alpha) \leq \frac{n^{n+1}\alpha^{n+1}}{n!} e^{-n\alpha}.$$

Using the Stirling formula for $n!$, we can compute an equivalent of this last expression:

$$\frac{n^{n+1}\alpha^{n+1}}{n!} e^{-n\alpha} \sim \frac{1}{\sqrt{2\pi}} e^{(n+1)\log\alpha + n(1-\alpha) + 1/2(\log n)}.$$

The function $u_n(\alpha) = [(n+1)\log\alpha + n(1-\alpha) + 1/2(\log n)]$ has a leading term in n equal to $n(\log\alpha + 1 - \alpha)$ which tends towards to $-\infty$ when n tends towards ∞. Then this expression tends towards 0 in the same conditions and also $I_n(\alpha)$.

$\alpha > 1$. The characteristics of the function $h(v)$ described above allow us to use the following lemma (Dieudonné, 1982):

Lemma. Let φ, h defined on $[a, b]$ such that: (1) φ, h are C^2 on $[a, b]$; (2) $\int_a^b |\varphi(x)| e^{h(x)}\, dx$ is defined; and (3) The derivative h' of h changes its sign at a unique point c, $c \in [a, b]$, which is a maximum for h, and such that $\varphi(c) \neq 0$ and $h''(c) < 0$. Then, for $n \to +\infty$, we have the following equivalence:

$$\int_a^b \varphi(x) e^{nh(x)}\, dx \sim \varphi(c) e^{nh(c)} \sqrt{\frac{2\pi}{-nh''(c)}}$$

When $\alpha > 1$, $c = 1/\alpha$ and this lemma can be applied to $J_n(\alpha)$, with $\varphi(v) = 1$ and $h(v) = \log v - \alpha v$. We have:

$$J_n(\alpha) \sim e^{n(-1-\log\alpha)} \sqrt{\frac{-2\pi}{n(-\alpha^2)}} = \frac{e^{-n}}{\alpha^{n+1}} \sqrt{\frac{2\pi}{n}}$$

and

$$I_n(\alpha) \sim \frac{n^{n+1}e^{-n}}{n!} \sqrt{\frac{2\pi}{n}} = \frac{n^n e^{-n}\sqrt{2\pi n}}{n!} \sim 1.$$

The last equivalence is obtained by the Stirling formula.

$\alpha = 1$. The same lemma can be used with the same result than in the case $\alpha > 1$, by integrating in an interval $[0, 1+\epsilon]$ and giving then to ϵ the value 0.

Conclusion

The series of functions f_n converges to a step function, evidently characteristic of a plug-flow reactor. We provide in Figure 2 the maximum relative error of a series of n well-stirred reactors with respect to a plug-flow reactor, depend-

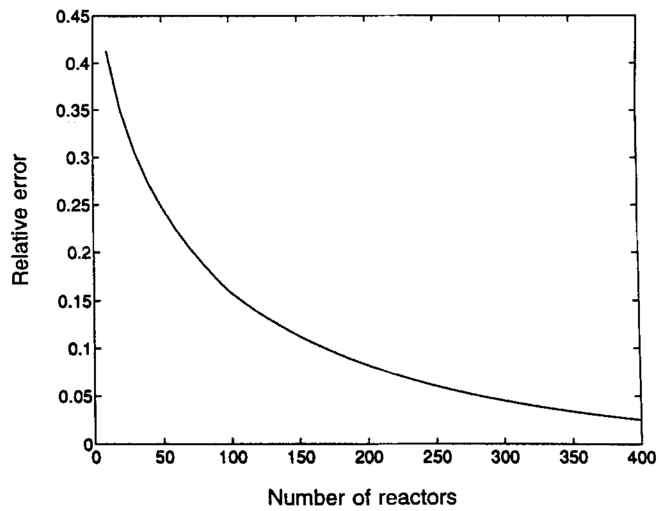

Figure 2. Relative error between a series of CSTR and the PFR.

ing on n, that is, the maximum difference between the step function with step at $t = 1$ and the function $f_n(t)$ ($t = 1$ was not a factor in the calculation). The convergence is so slow that for example for an enormous series of 400 reactors, the precision is only 97.5%.

Acknowledgment

We are grateful to Dr. L. Magnin for pointing to us the Lemma of Dieudonné.

Notation

C = internal and outlet concentration
C_0 = initial solute concentration
e = 2.71828182
$f_n = C_n/C_0$
i = index of the considered reactor
t = time variable
V = volume of the solution in the process
$\alpha = t/\tau$: reduced time
π = 3.14159265...
τ = time constant of a single reactor

Literature Cited

Dieudonné, J., *Calcul Infinitésimal*, Masson, Paris (1982).
Gervais, P., C. Bazelin, and A. Voilley, "Patterns of Aeration in a Solid Substrate Fermentor through the Study of the Residence Time Distribution of a Volatile Tracer," *Biotechnol. Bioeng.*, **28**, 1540 (1986).
Loncin, M., *Génie Industriel Alimentaire*, Masson, Paris (1976).
Moser, A., *Bioprocess Technology*, Springer-Verlag, New York (1981).
Villermaux, J., *Génie de la Réaction Chimique*, Tec et Doc, Lavoisier, Paris (1993).

Manuscript received Apr. 5, 1994 and revision received Sept. 12, 1994.