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Chemical Engineering Science 61 (2006) 4689–4696

Chemical Engineering Science

www.elsevier.com/locate/ces

A critical appraisal of the Π-criterion through continuation/optimization

G. D'Avino[a], S. Crescitelli[a], P.L. Maffettone[a], M. Grosso[b,*]

[a]*Dipartimento d'Ingegneria Chimica, Università "Federico II", Piazzale Tecchio 80, I-80125 Napoli, Italia*
[b]*Dipartimento di Ingegneria Chimica e Materiali, Università degli Studi di Cagliari, Piazza d'Armi I-09123, Cagliari, Italia*

Received 22 March 2005; received in revised form 9 February 2006; accepted 19 February 2006
Available online 6 March 2006

Abstract

Periodic forcing may improve bioreactor performances with respect to stationary operating conditions. An analytical procedure based on the so-called π-criterion was used by Parulekar [(1998). Analysis of forced periodic operations of continuous bioprocesses—single input variations. Chemical Engineering Science, 53(14), 2481–2502] to assess whether periodic forcing is beneficial, and, when this is the case, the criterion also gives an indication of the optimal forcing frequency. Such procedure is exact only in the limit of infinitesimal amplitude of the periodic forcing. In this work the applicability of the π-criterion to nonlinear models is investigated. The analysis is carried out by comparing the analytical predictions of the criterion with numerical predictions obtained with a continuation/optimization algorithm. As an example, two different reaction schemes are considered. The continuation/optimization procedure gives information on optimal forcing frequency and also on optimal forcing amplitude.

Keywords: Biochemical engineering; Dynamic simulation; Nonlinear dynamics; Numerical analysis; Optimization; Reaction engineering

1. Introduction

The analysis of periodically forced reactors has been widely addressed in the recent literature. The research is mainly aimed at assessing whether process performances could be improved by periodic forcing. Different forcing strategies have been developed: input variables are forced to vary in time; the reactor is forced by discontinuously inverting the flow direction; periodic forcing of reactor networks is obtained by cyclic permutation of the reactor order. The scientific interest was first focused to the characterization of fixed bed reactors operated with a periodical change of feed temperature and composition (e.g., Briggs et al., 1977). Reverse flow reactors proved useful in several heterogeneously catalyzed reactions (e.g., Matros, 1985; Matros and Bunimovich, 1996). Reactor networks with switched feed position were suggested as a possible alternative to reverse flow reactors (Haynes et al., 1992).

In this work, we are mainly interested in the determination of possible improvement in performance of continuous processes via periodic forcing of one process input. To this end, an oft-used approach is based on the so-called π-criterion (Abulesz and Lyberatos, 1987, 1989; Bittanti et al., 1973; Guardabassi et al., 1974; Parulekar, 1998, 2000, 2001, 2003; Sterman and Ydstie 1990a, 1990b; Watanabe et al., 1981). This criterion is an analytical tool valid for linear systems to ascertain whether periodic forcing of input variables may lead to performance improvements. It provides a sufficient (but not necessary) condition for this. In addition, the π-criterion also suggests the most favourable forcing frequency.

Parulekar (1998) thoroughly analyzed the performance of continuous stirred tank bioreactors (CSTBR) applying the π-criterion to an ample variety of kinetic schemes. That paper was a systematic analysis of the impact of forcing on biochemical processes. Parulekar pointed out that in order to apply the criterion to nonlinear systems the input variables should be subjected to small-amplitude forcing. As forcing amplitude increases, nonlinearities set in and the outcomes of the π-criterion may be in principle questionable.

* Corresponding author. Tel.: +39 0706755075; fax: +39 0706755067.
E-mail address: grosso@dicm.unica.it (M. Grosso).

0009-2509/$ - see front matter © 2006 Elsevier Ltd. All rights reserved.
doi:10.1016/j.ces.2006.02.024

A possible check on the validity of the π-criterion can be carried out by numerically analyzing the dynamic model. In this paper, we compare the results of the π-criterion for nonlinear kinetic schemes in CSTBR with those obtained with a numerical procedure based on a continuation/optimization algorithm. Of course, any numerical optimization algorithm can be adopted, we have chosen a continuation/optimization scheme as it is able to reconstruct the locus of optimal conditions as more parameter values are varied, automatically providing information on the optimal solution stability.

Parameter continuation is a typical tool to characterize bifurcations of nonlinear dynamical systems. It can be applied in optimization problems as the optimal conditions can be regarded as a critical point. Thus, when a maximum of an objective function is available, the parameter continuation algorithm automatically reconstructs the locus of optimal conditions as one or more parameters are varied with continuity. It so appears that this tool can be very useful in determining the loci of optimal conditions of frequency and amplitude of the forcing as one model parameter is varied. The continuation software here used is AUTO (Doedel et al., 1997).

For the sake of comparison, we have applied such a technique to study two cases of those examined by Parulekar (1998), namely the processes for the production of propionic acid with *Propionibacterium shermanii* and for the production of ethanol from glucose with *Saccharomyces cerevisiae*.

2. CSTBR model and kinetic schemes

The dynamics of the CSTBRs can be described within the assumptions of unstructured and unsegregated conditions by the mass balance equations for the biomass X, the substrate S and the product P

$$\begin{aligned}
\frac{dX}{dt} &= r_1 - DX, \quad r_1 = \mu X,\\
\frac{dS}{dt} &= D(S_F - S) - r_2, \quad r_2 = \sigma X,\\
\frac{dP}{dt} &= r_3 - DP, \quad r_3 = \varepsilon X.
\end{aligned} \tag{1}$$

In Eqs. (1), t is time, D is the so-called dilution rate, that is the reciprocal of the residence time; S_F is the substrate concentration of the feed; r_1 is the kinetic rate for growth of biomass; r_2 is the rate of consumption for the substrate, and r_3 is the kinetic rate for the formation of product. The parameters μ, σ, and ε are kinetic functions. Several expressions for these three rates are available in the literature. We here analyze the features of two kinetics already studied by Parulekar (1998). The first one is the kinetic scheme proposed by Ruan and Chen (1996) for the production of propionic acid with *Propionibacterium shermanii*

$$\begin{aligned}
\mu &= \frac{\mu_m S}{K_S + S + S^2/K_i}\left(1 - \frac{P}{P_m}\right)^n,\\
\sigma &= a\mu,\\
\varepsilon &= \alpha\mu + \beta.
\end{aligned} \tag{2}$$

In Eqs. (2) $\mu_m = 0.48\,\mathrm{h}^{-1}$, $P_m = 50\,\mathrm{g/l}$, $K_S = 1.2\,\mathrm{g/l}$, $K_i = 22\,\mathrm{g/l}$, $a = 2.5$, $\alpha = 2.2$, $\beta = 0.2\,\mathrm{h}^{-1}$, and $n = 1$ are kinetic parameters as given by Parulekar (1998), and their values have been kept constant. In the following, this kinetic scheme is referred as RC.

The second scheme is due to Aiba and Shoda (Aiba et al., 1968; Aiba and Shoda, 1969) and applies to the production of ethanol from glucose with *Saccharomyces cerevisiae*

$$\begin{aligned}
\mu &= \frac{\mu_m S}{(K_{S1} + S)}\,\mathrm{e}^{-\alpha_1 P},\\
\sigma &= a\mu,\\
\varepsilon &= \frac{\varepsilon_m S}{(K_{S2} + S)}\,\mathrm{e}^{-\beta_1 P}.
\end{aligned} \tag{3}$$

In Eqs. (3), $\mu_m = 0.408\,\mathrm{h}^{-1}$, $K_{S1} = 0.22\,\mathrm{g/l}$, $\alpha_1 = 0.028\,\mathrm{l/g}$, $a = 10$, $\varepsilon_m = 1.0\,\mathrm{h}^{-1}$, $K_{S2} = 0.44\,\mathrm{g/l}$, $\beta_1 = 0.015\,\mathrm{l/g}$ are kinetic parameters, the chosen constant values are, again, those given by Parulekar (1998). In the following, this kinetic scheme is referred as AS.

In the present paper, the forcing is imposed by modulating the substrate feed concentration. When an input is periodically forced, the dynamical system (1) can be formally written in the following form:

$$\frac{d\mathbf{x}}{dt} = \mathbf{F}(\mathbf{x}, \boldsymbol{\theta}, \mathbf{u}, t). \tag{4}$$

In Eq. (4), $\mathbf{x}$ is the vector of the state variables (i.e., $\mathbf{x} = [X, S, P]$, $\mathbf{x} \in R^3$), $\mathbf{F}$ is the periodic vector field, $\boldsymbol{\theta}$ is the vector grouping constant parameters, and $\mathbf{u}$ is the vector containing the parameters which fix the periodic input. Vector $\mathbf{u}$ can be considered as the manipulable input variable. The values of these parameters have to be chosen so as to optimize some objective function. Note that Eq. (4) can be used for unforced conditions as well, in that case $\mathbf{F}$ does not depend on $\mathbf{u}$, and is not an explicit function of time.

We are interested in determining whether periodic forcing could improve some reactor performance. In such a case, the optimal periodic forcing conditions (i.e., $\mathbf{u}$-values) have to be determined. For the sake of simplicity, we consider that the optimal conditions are those maximizing the reactor productivity. In the case of unforced reactor, and assuming that the stable solution is stationary steady state regime, the objective function J_{SS} is

$$J_{SS} = g(\mathbf{x}_{ss}, \boldsymbol{\theta}) = DP_{SS}. \tag{5}$$

In Eq. (5), $\mathbf{x}_{ss}$ represents the steady-state regime. Obviously, $\mathbf{x}_{ss}$ implicitly depends on $\boldsymbol{\theta}$, which may be chosen to maximize the productivity.

The productivity under unforced conditions is then compared with that obtained under periodic forcing. As already mentioned, we limit to the case of periodic forcing of the feed substrate concentrations, S_F, with the following sinusoidal waveform:

$$S_F = S_{F0}(1 + A\cos\omega t). \tag{6}$$

In passing, it should be remarked that Eq. (6) represents a very specific waveform for the forcing, and that different