Chemical Engineering Science 117 (2014) 71–84

Contents lists available at ScienceDirect

Chemical Engineering Science

journal homepage: www.elsevier.com/locate/ces

Nonlinear frequency response analysis of forced periodic operation of non-isothermal CSTR using single input modulations. Part I: Modulation of inlet concentration or flow-rate

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HIGHLIGHTS

- Evaluating single input periodic operations of non-isothermal CSTR by NFR method.
- Analysis for the non-isothermal, homogeneous, simple n-th order reaction in a CSTR.
- Derivation of asymmetrical second order FR functions and sign analysis.
- Conditions for process improvement by modulating inlet concentration or flow-rate.
- Comparison between results obtained by NFR method and by numerical simulations.

ARTICLE INFO

Article history:
Received 20 January 2014
Received in revised form
30 May 2014
Accepted 5 June 2014
Available online 14 June 2014

Keywords:
Non-isothermal CSTR
Nonlinear dynamics
Mathematical modelling
Chemical reactors
Simulation
Nonlinear frequency response method

ABSTRACT

Periodic operations of a non-isothermal CSTR with n-th order reaction, subject to a single input modulation, is analysed using the nonlinear frequency response (NFR) method, introduced in our previous publications. The method is based on deriving the asymmetrical second order frequency response function (FRF) and analysing its sign. In Part I of this paper, periodic operation with modulation of the inlet concentration or flow-rate of the reaction stream is analysed. As a result, conditions regarding the reaction order, process parameters and frequency of the input modulation are identified that need to be fulfilled in order to achieve process improvement through the periodic operation compared to conventional steady state operation. The method is applied for a numerical example from literature and the results obtained by the NFR method are compared with the results of numerical simulation. Good agreement is obtained, except for imposed forcing frequencies close to the resonant frequency and high forcing amplitudes.

1. Introduction

Periodic operation of different chemical engineering processes, especially chemical reactors, has been a research topic of a number of research groups in the last 50 years (Douglas and Rippin, 1966; Douglas, 1967; Horn and Lin, 1967; Renken, 1972; Bailey, 1973; Watanabe et al., 1981; Schadlich et al., 1983; Silveston 1987; Sterman and Ydstie, 1990a, 1990b, 1991; Chen and Hwang, 1994; Silveston et al. 1995; Silveston, 1998).

Periodic modulation of one or more inputs can provide better average performance compared to the optimal steady-state operation (increased conversion, improved selectivity, increased yield, increased catalytic activity etc.). The source of possible improvement lies in the process nonlinearity. Many experimental and simulation studies verify that it is often advantageous to exploit the nonlinear behaviour of chemical reactions and to operate in a dynamic regime by periodic modulation of one or more inputs (Sterman and Ydstie, 1991).

For nonlinear systems with periodic modulation one or more inputs, the average value of the output is different from the steady-state value. Although this difference is small for mild nonlinearities, for highly nonlinear systems or those which exhibit resonance, the deviations might be very significant (Douglas, 1967).

Identification of candidate systems for process enhancement through periodic operation and estimation of the magnitude of such enhancement have occupied many researchers. More details about previously proposed criteria or techniques for evaluation of periodically operated processes can be found in Petkovska and Seidel-Morgenstern (2013).

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http://dx.doi.org/10.1016/j.ces.2014.06.013
0009-2509/

These previous theoretical approaches have not provided yet general methods to predict the possibility of process improvements (Petkovska and Seidel-Morgenstern, 2013). In practice, testing whether a periodic operation leads to an increased productivity as compared to the corresponding steady-state operation, is usually performed by long and tedious experimental and/or numerical work. Therefore, there is still a need for developing simple and reliable methods which would enable quantitative evaluation of the possibility of process improvements through periodic operations (Petkovska and Seidel-Morgenstern, 2013).

In our previous work, we introduced the nonlinear frequency response (NFR) method, which can give in early development stages a fast answer whether working under periodic conditions could be favourable. The NFR method, which is applicable for weakly nonlinear systems (Weiner and Spina, 1980), is based on Volterra series, generalized Fourier transform and the concept of higher order frequency response functions (FRFs) (Marković et al., 2008). The NFR method also enables approximate evaluation of the magnitude of the improvement for weakly nonlinear systems if it can be achieved by periodic operation.

Till now, the NFR method has been applied for evaluation of periodic operations of different types of reactors (continuous stirred tank reactor (CSTR), plug flow tubular reactor and dispersive flow tubular reactor) with feed concentration modulation for simple isothermal homogeneous n-th order reactions (Marković et al., 2008). The same type of analysis was used for periodic operation of a CSTR with a simple isothermal heterogeneous catalytic n-th order reaction (Petkovska et al., 2010).

In one of our previous papers, the NFR method was extended for evaluation of periodic operation with simultaneous modulation of two inputs, and tested on an isothermal CSTR in which a simple isothermal n-th order homogeneous reaction takes place and inlet concentration and flow-rate are modulated simultaneously (Nikolić-Paunić and Petkovska, 2013).

In this paper, for the first time the NFR method is applied for evaluation of periodically operated non-isothermal reactors, for which the temperature effects of the chemical reaction cannot be neglected. This problem has already been treated in the literature (Ritter and Douglas, 1970; Sterman and Ydstie 1990a, 1990b; Dorawala and Douglas, 1971; Silveston and Hudgins, 2004). There are several parameters which could be periodically modulated: the inlet concentration, the flow-rate, the temperature of the feed stream and the temperature of the heating/cooling fluid. Since the potential for improvement through periodic operation strongly depends on the degree of the nonlinearity of the system, it is expected that the non-isothermal CSTR, which is highly nonlinear, would offer a lot of potential for process improvement. The non-isothermal CSTR is also a good test for the NFR method, considering that the method is valid for weakly nonlinear systems.

It is well-known that a non-isothermal CSTR can in principle exhibit unstable behaviour (Douglas, 1972). It should be noticed that the NFR method is applicable only for stable systems, so stability analysis should always be performed first.

In this paper, the NFR method is applied for evaluation of periodic operation of a non-isothermal CSTR in which a simple n-th order homogeneous reaction takes place, when either the inlet concentration and or the flow-rate are modulated inputs. In Part II of this paper, we will analyse the periodic operation of the non-isothermal CSTR when the modulated inputs are the temperature of the inlet reaction stream or the temperature of the cooling/heating fluid.

2. Nonlinear frequency response method for evaluating periodic processes

Frequency response (FR) represents a quasi-stationary response of the system to a periodic (sinusoidal or co-sinusoidal) input modulation, which is achieved when the transient response becomes negligible (theoretically for infinite time) (Douglas, 1972).

FR of a linear system is a periodic function of the same shape and frequency as the input, but with different amplitude and phase from the input values. The mean value of this periodic function is equal to the steady state value. Frequency response function of a linear system is defined by the amplitude ratio and the phase difference of the output and input in the quasi-stationary state (Douglas, 1972).

On the other hand, FR of a nonlinear system is a complex periodic function and it cannot be represented by a single frequency response function. FR of a nonlinear system, in addition to the basic harmonic, which has the same frequency as the input modulation, also contains a non-periodic, the so called DC component, and an infinite number of higher harmonics (Douglas, 1972; Weiner and Spina, 1980; Petkovska and Seidel-Morgenstern, 2013). One approach for analysing FRs of nonlinear systems is the concept of higher order frequency response functions (FRFs) which is based on Volterra series and the generalized Fourier transform (Weiner and Spina, 1980).

The nonlinear model $\boldsymbol{G}$ of a weakly nonlinear system in the frequency domain can be replaced by an infinite sequence of frequency response functions (FRFs) of different orders (Weiner and Spina, 1980). These FRFs are directly related to the DC component and different harmonics of the response (Weiner and Spina, 1980).

If the input is defined as a single harmonic periodic function with forcing amplitude A and forcing frequency ω

$$x(t) = x_s + A\cos(\omega t) \tag{1}$$

for infinite time, the output of a weakly nonlinear system is obtained as a sum of a DC component and the first, second, … harmonics

$$\begin{aligned} y(t) &= y_s + y_{DC} + y_I + y_{II} + \dots \\ &= y_s + y_{DC} + B_I\cos(\omega t + \varphi_I) + B_{II}\cos(2\omega t + \varphi_{II}) + \dots \end{aligned} \tag{2}$$

The DC component, which is responsible for the time-average performance of periodic processes and which is most essential in this paper, can be expressed as the following infinite series (Weiner and Spina, 1980):

$$y_{\mathrm{DC}} = 2\left(\frac{A}{2}\right)^2 G_2(\omega, -\omega) + 6\left(\frac{A}{2}\right)^4 G_4(\omega, \omega, -\omega, -\omega) + \dots \tag{3}$$

where $G_2(\omega, -\omega)$ represents the asymmetrical second order frequency response function, $G_4(\omega,\omega,-\omega,-\omega)$ the asymmetrical fourth order FRF, etc.

For weakly nonlinear systems, the contributions of the higher other FRFs decrease with the increase of their order (Petkovska and Seidel-Morgenstern, 2013).

The dominant term of the DC component is proportional to the asymmetrical second order FRF, $G_2(\omega,-\omega)$ and the approximate value of the DC component can be easily calculated from (Marković et al., 2008)

$$y_{DC} \approx 2\left(\frac{A}{2}\right)^2 G_2(\omega, -\omega) \tag{4}$$

In this way, the sign of the asymmetrical second order FRF $G_2(\omega,-\omega)$ defines the sign of the DC component. Consequently, in order to decide whether a particular periodic operation is favourable compared to the optimal steady-state operation, it is enough to derive and analyse the second order asymmetrical FRF (Marković et al., 2008). It is also possible to calculate approximately the magnitude of the improvement, by estimating the value of the second order asymmetrical FRF for chosen values of the forcing amplitude and forcing frequency.