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Finding the best strategy for periodic operation of a CSTR with flow-rate modulation – The nonlinear frequency response approach

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Highlights

- Periodic operation of an isothermal CSTR with flow-rate modulation was analysed using the nonlinear frequency response method.
- 3 strategies were investigated: 1. Both flow-rates (in and out) are modulated in the same way; 2. Only the inlet flow-rate is modulated; 3. Only the outlet flow-rate is modulated.
- For each case (reaction order and *Da* number) a strategy that improves reactor performance has been found.

1. Introduction

It has been known for quite some time that one way to intensify a chemical reactor can be through periodic operation, by forced periodic modulations of one or more of its inputs [1]. The intensification is a result of the system non-linearity. Nevertheless, finding out whether, at which conditions and to which extent the periodic operation can be superior to the optimal steady-state one is a serious challenge. One approach that can be used is an approximate, analytical method called nonlinear frequency response (NFR) method [2]. The NFR method is relatively new, fast and easy to apply and it can be used to evaluate the performance of forced periodically operated chemical reactors. It is based on the concept of higher order frequency response functions (FRFs) and applicable for stable weakly nonlinear systems [2]. Frequency response of a weakly nonlinear system, in addition to the periodic terms, contains a non-periodic term, which is responsible for the time-average performance of the periodically operated reactor. Its sign and value define whether, and to which extent, the periodic operation leads to process improvement. Using the NFR method, this non-periodic term can be estimated from a single asymmetrical second order (ASO) FRF (for single input modulation).

An input that can easily be modulated periodically is a flow-rate of a stream that enters or leaves the reactor. In our previous work, we investigated the influence of the flow-rate modulations on isothermal [3] or a non-isothermal [4] CSTRs with a simple reaction mechanism (A→P), under constant volume conditions (when the inlet and outlet flow-rate are changing synchronically). Nevertheless, other modes of modulating the flow-rates are also possible, e.g. modulating only the inlet, or only the outlet flow-rate. In these cases, the volume of the CSTR is also changing periodically. This paper is devoted to finding the best strategies of the flow-rate modulation for different cases, so the best one can be chosen for a particular case. The analysis is performed for an isothermal CSTR with a simple reaction mechanism A→P.

2. Methods – deriving the ASO FRFs

The NFR method is based on derivation and sign analysis of the ASO FRF correlating the output and input of interest. The starting point in this derivation is setting up the appropriate mathematical model. The general mathematical model of an isothermal CSTR with flow-rate modulation can be defined by a material balance of the reactant A and the total material balance. If dimensionless input and output variables are introduced, defined as relative deviations from the corresponding steady-state values, these equations can be written in the following way:

$$\frac{dV}{d\tau}+\frac{dC_A}{d\tau}+V\frac{dC_A}{d\tau}+C_A\frac{dV}{d\tau}=(1+Da)(1+\Phi_i)-(1+\Phi)(1+C_A)-Da(1+V)(1+C_A^n);\qquad \frac{dV}{d\tau}=\Phi_i-\Phi$$

where V, C_A, Φ and Φ_i are, consecutively, dimensionless reactor volume, concentration of A, outlet and inlet flow-rate, τ is dimensionless time, n the reaction order and Da is the so-called Damköhler number. Starting from these model equations and following a standard derivation procedure [2], the ASO FRFs correlating the outlet molar flow rate of A and the modulated flow-rate were derived for the following 3 different strategies:

Strategy 1. $\Phi(\tau)=\Phi_i(\tau)=A\cos(\omega\tau)$ (V=const)

Strategy 2. $\Phi_i(\tau)=A\cos(\omega\tau)$,$\Phi$=const

Strategy 3. $\Phi(\tau)=A\cos(\omega\tau)$,$\Phi_i$=const

The ASO FRFs for all 3 cases are functions of the dimensionless frequency ω, with n and Da as parameters. According to the NFR method [2], the periodic operation can be expected to be favourable if the corresponding ASO FRF corresponding to the molar flow-rate of the reactant is negative.

3. Results and discussion

A summary of the sign analysis of the ASO FRFs corresponding to the 3 different strategies is given in Table 1. Obviously, Strategy 1 can be beneficial only for negative reaction orders, while Strategies 2 and 3 can bring improvements both for positive and negative values of n, by choosing the appropriate frequency of the input. (For n=0, no improvement is possible, as the system is essentially linear.). It needs to be pointed out that in Strategies 2 and 3 the volume of the reaction mixture also changes periodically and the amplitude of the flow-rate modulation has to be limited in such a way that the volume stays in its physical boundaries.

Table 1. Overview of applicability of the 3 different strategies of flow-rate modulation

Reaction order	Strategy	Conditions
$n>0$	1	Never
	2	$0<n<1$ for $Da<\frac{2}{1-n}$ and $\omega^2>\frac{(n+1)Da+2}{(n-1)Da+2}$
		$n\geq1$ for $\forall Da$ and $\omega^2>\frac{(n+1)Da+2}{(n-1)Da+2}$
	3	$\forall Da$ and $\omega^2>(n+1)Da+2$
$n=0$	1, 2 and 3	Never
$n<0$	1	$\forall Da$ and $\forall\omega$
	2	$-1\leq<n<0$ for $Da>\frac{2}{1-n}$ and $\forall\omega$
		$n<-1$ for $\begin{cases} Da<\frac{2}{1-n} \text{ and } \omega^2<\frac{(n+1)Da+2}{(n-1)Da+2} \\ \frac{2}{1-n}<Da<-\frac{2}{1+n} \text{ and } \forall\omega \\ Da<-\frac{2}{1+n} \text{ and } \omega^2>\frac{(n+1)Da+2}{(n-1)Da+2} \end{cases}$
	3	$-1\leq<n<0$ for $\forall Da$ and $\forall\omega$
		$n<-1$ for $Da>-\frac{2}{1+n}$ and $\omega^2>(n+1)Da+2$

4. Conclusions

The analysis based on the NFR method enables finding the best strategy of modulating the inlet and/or outlet flow-rate in an isothermal CSTR, so that the periodic operation is superior to the corresponding steady-state one. Which strategy is the best depends preferably on the reaction order n and the Da number. It is important to point out that a beneficial strategy can be found for all cases except for n=0.

References

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Keywords

Periodic operation; CSTR; Flow-rate modulation; Nonlinear Frequency Response