

Forced periodic reactor operation with simultaneous modulation of two inputs: Experimental investigation based on Nonlinear Frequency Response Analysis

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Highlights

- Using the nonlinear frequency response (NFR) method to evaluate theoretically the potential of perturbing simultaneously two inlets to improve reactor performance
- Implementation of the NFR for hydrolysis of acetic anhydride carried out as a model reaction in a lab-scale CSTR

1. Introduction

The classical design of continuous chemical reactors assumes that they are operated in a steady-state which is usually optimized and maintained by using an appropriate control system. Nevertheless, it has been known for quite some time that, in some cases, better performance can be achieved by applying a periodic regime, by forced periodic modulations of one or more inputs to the reactor [1,2]. Finding out whether, at which conditions and to which extent the periodic operation can be superior to the optimal steady-state is a serious challenge. One approach that can be used is an approximate, analytical method called nonlinear frequency response (NFR) method [3]. In this work we use the NFR method in order to find conditions in which a CSTR with a test reaction of acetic acid anhydride hydrolysis can be run in periodic mode which would be superior to the corresponding steady-state operation. Based on the results of the NFR analysis, experimental investigation is performed using a lab-scale CSTR.

2. Methods

The nonlinear frequency response (NFR) method is a relatively new, fast and easy to apply analytical method for evaluating the performance of forced periodically operated chemical reactors. It is based on the concept of higher order frequency response functions (FRFs), which is applicable for weakly nonlinear systems [3]. Frequency response of a weakly nonlinear system, in addition to the basic harmonic, contains a non-periodic (DC) term and, theoretically, an infinite sequence of higher harmonics. The DC component of the output is responsible for the average performance of the periodically operated reactor, and its sign and value define whether, and to which extent, the periodic operation leads to process improvement. Using the NFR method,

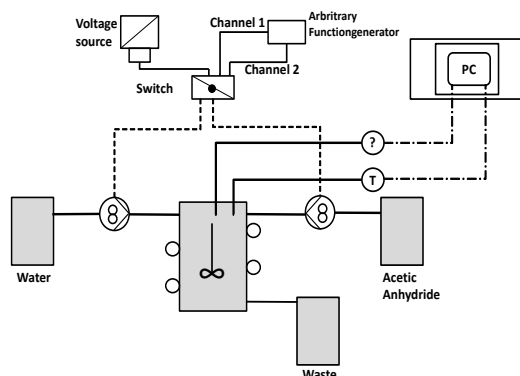


Fig. 1. Experimental setup for testing periodic operation for acetic anhydride hydrolysis

this DC component can be approximately estimated from a single asymmetrical second order FRF (for modulation of a single input) or from several single input and cross-asymmetrical second order FRFs (for multiple-input modulation). For the case of multiple modulated inputs, the optimal phase difference between the modulated inputs, which is an essential parameter, can be directly determined [4,5]. Promising parameters to be periodically modulated separately or simultaneously are clearly the reactant inlet concentrations, the flow-rates and the feed temperatures. The original method was developed for sinusoidal forcing functions. Recently it was extended to any shape of periodic input profile, e.g. square-wave [8].

The experimental system is shown in Fig. 1. A lab-scale reactor is fed with two input streams (water and acetic anhydride) with adjustable flow-rates, which enables modulation of the inlet reactant concentration or/and total flow-rate in the desired manner. The acetic acid concentration and temperature in the reactor are measured online and used to monitor the process performance.

3. Results and discussion

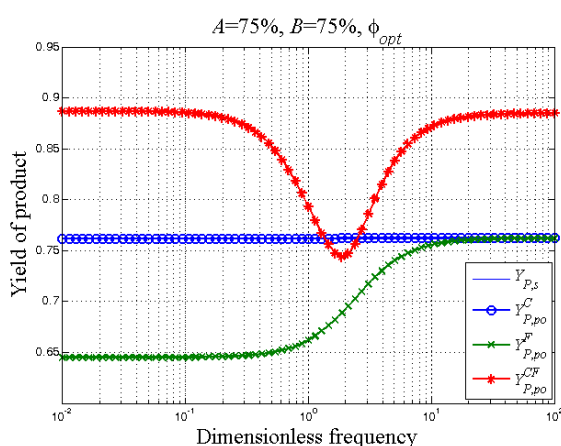


Fig. 2. Product yield of the periodic operation predicted by the NFR analysis: blue -inlet conc. modulation; green - flow-rate modulation; red – simultaneous modulation of both inputs

Based on theoretically developed FRFs for an adiabatic CSTR [5], the NFR analysis was performed for the investigated model reaction: acetic anhydride hydrolysis, using the kinetic data obtained in preliminary experimental investigation. Optimization of the steady-state operation of the reactor was performed first (taking into account physical constrains of the system), and periodic operations around that steady-state were analysed and compared to the optimal steady-state operation. Product (acetic acid) yield was used for comparison.

The NFR analysis showed that periodic modulation of only the inlet acetic anhydride concentration has practically no effect on the product yield, while only total flow-rate modulation results with lower yields than the corresponding steady-state operation. Nevertheless, simultaneous modulation of both inputs, with an appropriate phase difference, can results with substantial improvement, for low or for high forcing frequencies. These results, for amplitudes of both inputs of 75 % and for the optimal phase difference, are shown in Fig. 2. The NFR analysis also showed that best results would be obtained for square-wave modulation of the inputs.

Based on these results, a systematic experimental investigation was performed. By adjusting the flow-rates of the two feed pumps shown in Fig. 1. the inlet concentration and the total flow rate were simultaneously modulated with the same frequency and the corresponding optimal phase difference. According to the NFR results shown in Fig. 2, only the dimensionless frequencies below 0.1 and over 20 were used. The experimental results confirmed the predictions of the NFR analysis.

4. Conclusions

The experimental investigation confirmed the predictions of the NFR analysis, that modulating the inlet acetic anhydride concentration and the total flow-rate in the CSTR with appropriate frequency and phase difference can increase the yield of the acetic acid substantially.

Keywords

Forced periodic operation of reactors, perturbing simultaneously two inlets, nonlinear frequency response method.

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