Forced periodic reactor operation with simultaneous modulation of two inputs:

Experimental investigation based on Nonlinear Frequency Response Analysis

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Highlights

- Theoretical evaluation of possible reactor improvement for periodic operation.
- Automated experimental setup was built, with a lab-scale CSTR and adjustable flow-rates, in which periodic operations can be conducted.
- The concept was implemented for the hydrolysis of acetic anhydride as a model reaction.

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 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].

A challenging task is the identification and evaluation of superior periodic operation conditions. One approach that can be used is an approximate, analytical method called nonlinear frequency response (NFR) method [3]. The focus of this work is the experimental verification of results obtained using the NFR method. The conditions were chosen to perform the acetic anhydride hydrolysis as a test reaction in a CSTR exploiting a periodic operation mode which is superior to the corresponding steady-state operation.

2. Methods

The Nonlinear Frequency Response (NFR) method is a relatively new, fast and easy to apply analytical method, capable of predicting the performance of forced periodically operated chemical reactors. The method is based on the concept of higher order frequency response functions (FRF) and it 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 directly linked to the 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 DC component can be approximated 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 for periodic modulation are the inlet concentration, the total flow rate and the feed temperature. The original method was developed for a sinusoidal forcing function. Though, it can be extended to any shape of periodic input, such as square-wave [6].

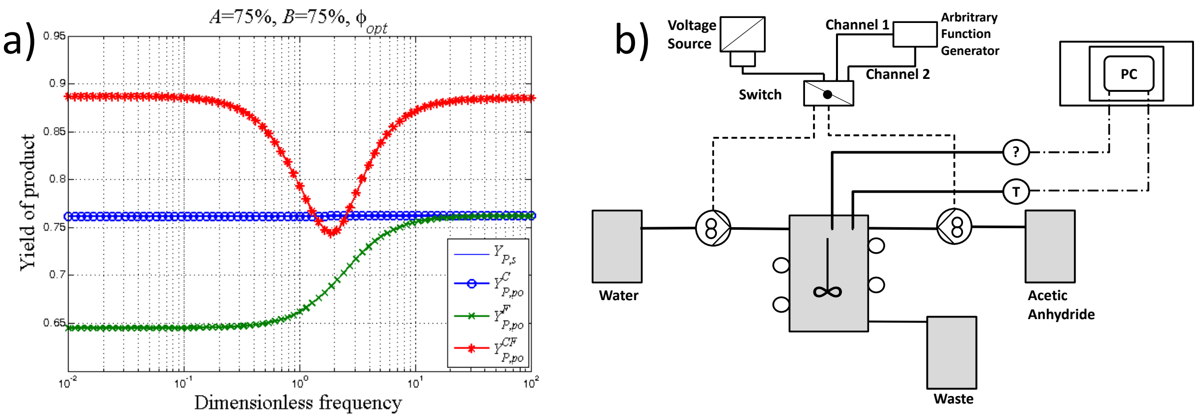

Fig. 1. a) Product yield of the periodic operation predicted by the NFR analysis: blue -inlet conc. modulation; green - flow-rate modulation; red – simultaneous modulation of both inputs for the experimentally developed setup for the hydrolysis of acetic anhydride, shown in b).

3. Results and discussion

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 investigations. Optimization of the steady-state operation of the reactor was performed first, taking into account physical constrains of the laboratory setup. Periodic operations around a distinct steady-state were analyzed and compared to the optimal steady-state operation. Product (acetic acid) yield was used for comparison, since it can be monitored using conductivity measurements. 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 A=B=75 % and for the optimal phase difference φ_{opt}, are shown in Fig. 1a). 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, using a specifically designed experimental setup, suitable to implement varying periodic function. By adjusting the flow-rates of the two feed pumps shown in Fig. 1b). the inlet concentration and the total flow rate were simultaneously modulated with the same frequency and the corresponding optimal phase difference.

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 well selected frequencies and phase differences can increase the yield of acetic acid substantially.

References

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Keywords
"nonlinear frequency response method", "forced periodic operation of reactor", "experimental validation", "perturbing simultaneously two inlets"