Forced periodic reactor operation: Analysis of process and forcing parameters exploiting the Nonlinear Frequency Response Method

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Continuous operation of chemical reactors is typically based on exploiting steady state conditions. However, theoretical investigations have shown that a forced periodic unsteady operation can generate higher conversion and yield, respectively [1,2]. Such concepts are obviously more complex and the outcoming information is difficult to predict, though a necessity. A powerful analytical approach to analyse and optimize periodic systems is the nonlinear frequency response (NFR) method [3].

The presented work is focused on providing theoretical and experimental results devoted to demonstrate both the potential of forced periodic operation and the strength of the NFR method to identify suitable operating conditions. The hydrolysis of acetic anhydride is studied experimentally as a model reaction applying an adiabatic continuous stirred tank reactor (CSTR). Key features of the investigations are the exploitation of two simultaneously modulated inputs and the evaluation of the effect of the input shapes. Figure 1 illustrates the considered operation modes for different input modulations. An effective operation should improve a certain objective Y with respect to the standard steady state operation.

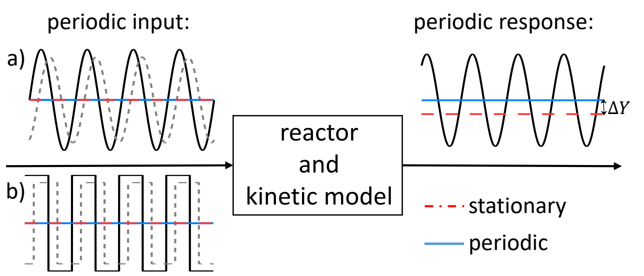

Figure 1:
Schematic illustration of double input modulation with a phase shift for: a) sinusoidal and b) square wave functions. ΔY describes the change of a performance criterion with respect to steady state operation.

The simultaneously modulated inputs are the anhydride concentration and the volumetric flow-rate. To maximize the performance these two inputs are modulated using the same forcing frequency. The reactor performance and possible improvements are strongly affected by another degree of freedom, namely the phase difference φ between two modulated inputs. Provided a model is available for the reaction kinetics, the NFR method can be applied to identify an optimal phase shift to maximize, for example, the time-average molar flow of the product [4,5]. Hereby, uncertainties in the kinetic, thermodynamic and process parameters have an influence on the predicted output of the dynamic reactor operation. A corresponding sensitivity analysis was done, which reveals the importance of correct kinetic parameters for a reliable prediction of the process performance.

To validate the predictions, an experimental set-up has been developed consisting of a double-jacketed glass reactor. Crucial is in particular a precise control of the fluctuating inlet streams of water and anhydride. To achieve this, an automated set-up is equipped with a function generator. The also fluctuating conversion can be measured with high time resolution using a conductivity sensor.

The application of optimal forcing parameters led to significant performance improvements. In very good agreement with the prediction of the NFR method the optimal phase difference between two sinusoidal inputs was found for φ approximately 150° (Figure 2). Further improvements are possible by modifying the input shapes from sinusoidal to square waves (see Figure 1).

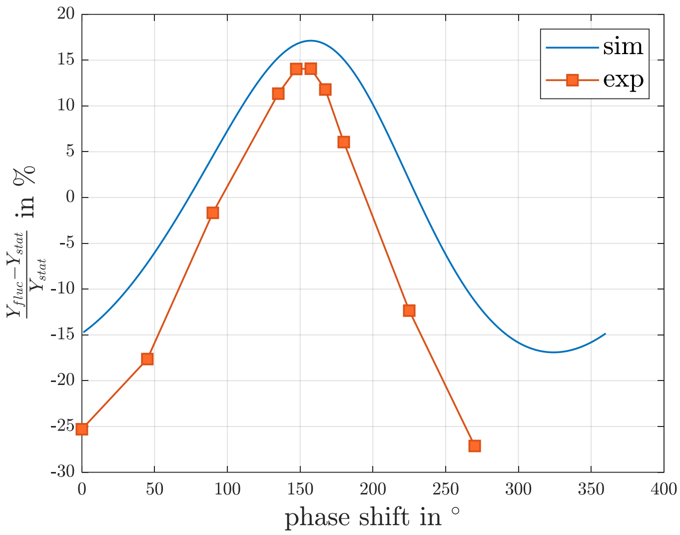

Figure 2:
Normalized difference of product yield
(simultaneous modulation of two inputs as sinusoidal shape)
Parameters:

Amplitudes:	$A_{Conc.} = 0.85$, $A_{Flow} = 0.55$
Stationary input parameters:	$T^{in} = 22°C$, $V_R = 298$ ml, period time = 40 min
Stationary input parameter, which are periodically modulated around the steady state values:	$c_A^{in} = 0.74 \frac{mol}{L}$ in water, $\dot{V}_{total} = 43$ ml/min

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