Forced periodic reactor operation

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The classical design of continuously operated chemical reactors assumes that they are operated in a steady-state which is usually optimized and maintained by using appropriate control systems. Nevertheless, it has been known for quite some time that, in some cases, better performance can be achieved by applying a periodic regime exploiting forced modulations of one or more inputs to the reactor [1,2]. Finding out whether, at which conditions and to which extent periodic operation can be superior to the optimal steady-state is difficult. One approach that can be used is an approximate, analytical method called nonlinear frequency response (NFR) method [3].

The NFR method is based on the concept of higher order frequency response functions (FRFs) and 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, 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 the reactant inlet concentrations, the flow-rates and the feed temperatures.

We used the NFR method in order to identify forced periodic conditions under which the acetic acid anhydride hydrolysis (chosen as a test reaction) can be favorably performed in a CSTR. Based on the results of the theoretical analysis, experimental investigations were performed using a lab-scale reactor exposed to two fluctuating inlet streams (water and acetic anhydride) with adjustable flow-rates, which enables modulation of the inlet reactant concentrations or/and total flow-rates in a flexible manner. The concentration of acetic acid formed is measured in the reactor online and used to monitor the process dynamics. Averaged values of the product outlet stream serve to validate the mean values predicted by NFR analysis and to evaluate the potential of this flexible forcing strategy.

Recently we started to analyze both theoretically and experimentally the potential of applying a forced periodic operation to improve the methanol synthesis from CO, CO_2 and H_2 using the conventional $Cu/ZnO/Al_2O_3$ catalysts. This work is based on a recently published detailed model of this reaction, which is capable to quantify the rates under dynamic conditions [6].

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