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APPLICATION OF NON-LINEAR FREQUENCY RESPONSE METHOD FOR INVESTIGATION OF PERIODICALLY OPERATED CHEMICAL REACTORS

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ABSTRACT. Process intensification aims to increase the efficiency of processes in the chemical industry by development of new concepts of processing methods and equipments. One way to achieve better performance is to operate the process in a periodic way. Periodic operation is achieved by periodic modulation or periodic forcing of one or more inputs into the system around a chosen steady-state point. For nonlinear systems the average value of the output when the input is periodically modulated is different from the steady-state value. For some cases, the periodic process can be superior to the optimal steady-state operation [2], while for some other it gives worse results. Most chemical processes are nonlinear in nature, and consequently they are candidates for possible improvements (increased conversion, selectivity, yield, production rates) through periodic operation.

Evaluating whether a periodic operation of a chemical processes would be beneficial and selecting the optimal operational conditions that maximize the process performance is often carried out by experimental and/or numerical studies. However, the experimental approach, as well as numerical simulations, can be rather time consuming and costly. It is therefore of economic importance to carry out theoretical studies alternatively for assessing the effect of periodic operation of a chemical processes and then selecting the optimal forcing inputs.

We will present a new, fast and easy method for evaluation of system performance in periodic operation, called the nonlinear frequency response method (NFRM). Nonlinear frequency response is a quasi-stationary response of a nonlinear system to a periodic (sinusoidal or co-sinusoidal) input change, around a steady-state. One of the most convenient tools for treating nonlinear frequency responses is the concept of higher order frequency response functions (FRFs), which is based on Volterra series and generalized Fourier transform. This concept is very convenient for analysing weakly nonlinear systems [1].

Frequency response of a weakly nonlinear system, in addition to the basic harmonic, contains a non-periodic (DC) term, and an indefinite sequence of higher harmonics. A nonlinear model with polynomial nonlinearity ($\boldsymbol{G}$), can be replaced by an indefinite sequence of FRFs of different orders ($G_1(\omega_1), G_2(\omega_1, \omega_2), G_3(\omega_1, \omega_2, \omega_3), \dots$), which are directly related to the DC component and different harmonics of the response. It is important to notice that the DC component is responsible for the average performance of the periodic process. On the other

hand, the DC component has a dominant term which is proportional to the asymmetrical second order FRF $G_2(\omega,-\omega)$, so it can be roughly estimated based on this function. The sign of the asymmetrical second order FRF $G_2(\omega,-\omega)$, defines the sign of the DC component and, consequently gives an answer to the question whether the periodic operation is favourable or not.

The NFRM can be extrapolated to nonlinear systems with multiple modulated inputs. To define the model with multiple modulated inputs, it is necessary to define several sets of FRFs which correlate each output to each input and cross-FRFs which correlate each output to several inputs.

In our previous work [3, 4], the NFRM was used to analyze the periodic performance of three standard reactor types: continuous stirred tank reactor, a plug flow tubular reactor (PFTR), and a dispersive flow tubular reactor (DFTR) with periodic changes of the input concentration. The cases of homogeneous [3] and heterogeneous [4] *n*-th order reaction takes place under isothermal conditions.

This method is also applied for analysis of periodic reactor operation for: 1) isothermal CSTR in which simple *n*-th order homogeneous reaction takes place, when inlet concentration and flow are modulated simultaneously and separately, 2) non-isothermal CSTR for simple *n*-th order homogeneous reaction when inlet concentration and temperature of inlet stream are modulated separately and simultaneously .

It has been estimated that conversion in chemical reactors can be significantly improved by periodic operation, for certain values of the reaction order (*n*) and for some modulated inputs, forcing parameters (periods, amplitudes, phase shift between synchronized inputs). The results obtained by nonlinear frequency response method were compared with the results of numerical solutions, and good agreement was obtained.

Keywords: nonlinear frequency response method, frequency response functions, periodic operation, chemical reactor

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