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Optimal design of the inlet temperature based periodic operation of non-isothermal CSTR using nonlinear output frequency response functions

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Abstract: The periodic operation of a non-isothermal continuous stirred tank reactor (CSTR) using inlet temperature modulation is investigated in this paper. The DC component of the CSTR output concentration is optimized by tuning the modulation parameters of the inlet temperature in order to achieve a maximum conversion using a Nonlinear Output Frequency Response Functions (NOFRFs) based approach. The results show that the new approach is fast and efficient in the analysis and design of the periodic operation of CSTR and can potentially be applied to conduct the optimal design of periodic operation of other chemical engineering processes.

Key words: optimal design, periodic operation, nonlinear output frequency response functions (NOFRFs), frequency domain

1. INTRODUCTION

Periodic operations in chemical engineering processes have received extensive attentions in the past decades (Bailey, 1973; Silveston et al., 1995; Petkovska et al., 2010). The advantages of the periodic operations lie in the fact that the average performance of a nonlinear chemical engineering system under a periodic operation is often superior to the steady-state performance under conventional constant input operations (Douglas, 1972). Chemical reactors under the condition of a periodic modulation feed were studied both theoretically and experimentally (Silveston et al., 2012;Brzić et al. 2015). In these chemical reactors, the CSTR is a good candidate because of its significant nonlinearity and has therefore been widely used to study the periodic operation of the flow-rate, concentration or temperature for the improvement of the process operational performance.

In recent years, the periodic operation of the CSTR has been investigated using nonlinear frequency analysis. Particularly, the generalized frequency response functions (GFRFs) method has been applied for the analysis of different types of CSTR for enhancement of process performance through periodic modulation of single or multiple inputs (Marković et al.,2008; Nikolić et al., 2014, 2015). However, the GFRFs method requires derive the higher order frequency response functions (FRFs), which is difficult to be implemented and widely applied in practice.

The NOFRFs is a novel concept for the analysis of nonlinear systems in the frequency domain (Lang et al., 2007). The method allows the analysis of nonlinear systems to be implemented in a manner similar to the analysis of linear systems and provides great insight into the mechanisms underlying many nonlinear behaviors (Peng, et al., 2007). In the present study, the NOFRFs method is applied for the analysis and design of a non-isothermal CSTR under a periodic operation of the inlet temperature. The results demonstrate the advantage of the new approach over existing GFRFs based analysis and the potential of the new analysis in the design of periodic operations for chemical engineering processes.

2. ISSUES ASSOCIATED WITH ACHIEVING A FAVORAVLE PERIODICAL OPERATION

The output response of nonlinear systems that are stable at zero equilibrium can be represented by a Volterra series (Rugh, 1981):

$$y(t) = y_s + \sum_{m=1}^{\infty} y_m(t) \tag{1}$$

where y_s is the steady-state output corresponding to a steady-state input, $y_1(t)$ is the system linear response, and, $y_m(t), m \geq 2$ is the system *m*th order nonlinear response.

When the system is subject to a harmonic input around a steady state x_s with amplitude A and frequency ω_F

$$x(t) = x_s + A\cos(\omega_F t) \tag{2}$$

Eq.(1) can be expressed as (Douglas, 1972)

$$y(t) = y_s + y_{DC} + B_1\cos(\omega_F t + \varphi_1) + B_2\cos(2\omega_F t + \varphi_2) + \ldots \tag{3}$$

where B_n and $\varphi_n, n = 1, 2$ are the amplitude and phase of the first and second order harmonic output of the system, respectively. y_{DC} is the DC component of the output response produced by the system nonlinearity which can be expressed as follows (Weiner and Spina,1980)

$$\begin{aligned} y_{DC} &= 2\left(\frac{A}{2}\right)^2 H_2(\omega_F,-\omega_F) \\ &+6\left(\frac{A}{2}\right)^4 H_4(\omega_F,\omega_F,-\omega_F,-\omega_F)+\cdots \end{aligned} \tag{4}$$

where $H_{2n}(\cdot), n=1,2,...$ represents the $2nth$ order GFRFs of the system.

Considering a chemical reaction

A→product(s)

where one or more inputs can be modulated periodically around an established steady-state. If this type of chemical reaction process is subject to the harmonic input (2) and the outlet concentration of the reaction can be represented by Eq (3). Marković et al. (Marković et al., 2008) have shown that the mean of the outlet concentration denoted by $c_A^m = y_s + y_{DC}$ is different from the corresponding steady state outlet concentration denoted by $c_{A,s} = y_s$ and the difference $\Delta = c_A^m - c_{A,s} = y_{DC}$ is the indicator of the process improvement that can be achieved by the introduction of a period operation. If $\Delta = y_{DC} < 0$, the introduction of the periodic operation is favorable, as it increase the conversion in comparison to the steady state operation. Otherwise, i.e. when $\Delta = y_{DC} > 0$, the periodic operation is unfavorable. Consequently, in order to introduce a favorable periodic operation, it is necessary to ensure that $\Delta = y_{DC} < 0$. Theoretically, this can be achieved using Eq.(4) but the multi-dimensional nature of the GFRFs $H_{2n}(\cdot), n=1,2,...$ in Eq.(4) and associated complexities imply that this is difficult in practice. Currently, the solution is to assume that Eq.(4) can be approximated well by the first term and analytically evaluate the sign of $H_2(\omega,-\omega)$ using the physical model of the CSTR to assess whether a favorable periodic operation is achievable. There are two fundamental problems with this available solution. First, it is impossible or difficult to analytically derive and study $H_2(\omega,-\omega)$ if the physical model of CSTR is not available or complicated. Secondly, this approach cannot be used to optimally determine the periodic operation parameters A and ω to minimize $\Delta = y_{DC}$ so as to maximize the conversion. In the present study, this challenge will be addressed by the development of a new approach based on the concept of Nonlinear Output Frequency Response Functions (NOFRFs).

3. THE NOFRFS METHOD

3.1 The NOFRFS concept

For system (1), Lang and Billings (Lang et al., 2007) have derived an expression for the output frequency response

$$\begin{cases} Y(j\omega) = \sum_{n=1}^{N} Y_n(j\omega) \quad for\ \forall\omega \\ Y_n(j\omega) = \dfrac{1/\sqrt{n}}{(2\pi)^{n-1}} \int_{\omega_1+\cdots+\omega_n=\omega} H_n(j\omega_1,\cdots,j\omega_n) \\ \qquad \times \prod_{i=1}^{n} U(j\omega_i) d\sigma_{n\omega} \end{cases} \tag{5}$$

where N is the maximum order of system nonlinearity, $Y(j\omega)$ and $U(j\omega)$ are the system input and output spectrum, $Y_n(j\omega)$ represents the n th order output frequency response and

$$\begin{aligned} H_n(j\omega_1,\cdots,j\omega_n) = \int_{-\infty}^{\infty}\cdots\int_{-\infty}^{\infty} h_n(\tau_1,\cdots,\tau_n) \\ \times e^{-(\omega_1\tau_1+\cdots+\omega_n\tau_n)j} d\tau_1\cdots d\tau_n \end{aligned} \tag{6}$$

is the definition of the *n*th order GFRFs with

$$\int_{\omega_1+\cdots+\omega_n=\omega} H_n(j\omega_1,\cdots,j\omega_n) \prod_{i=1}^{n} U(j\omega_i) d\sigma_{n\omega} \tag{7}$$

denoting the integration of $H_n(j\omega_1,\cdots,j\omega_n)\prod_{i=1}^{n} U(j\omega_i)$ over the n-dimensional hyper-plane $\omega_1+\cdots+\omega_n=\omega$.

Based on Eq.(5), the concept of the NOFRFs of nonlinear system (1) was introduced by Lang and Billings (Lang and Billings,2005) as

$$G_n(j\omega) = \frac{Y_n(j\omega)}{U_n(j\omega)}, n=1,\ldots,N \tag{8}$$

where

$$U_n(\mathrm{j}\omega) = \frac{1/\sqrt{n}}{(2\pi)^{n-1}} \int_{\omega_1+\cdots+\omega_n=\omega} \prod_{i=1}^{n} U(\mathrm{j}\omega_i) \mathrm{d}\sigma_{n\omega} \tag{9}$$

Clearly, using the NOFRFs concept, Eq.(5) can be written as

$$Y(j\omega) = \sum_{n=1}^{N} Y_n(j\omega) = \sum_{n=1}^{N} G_n(j\omega) U_n(j\omega) \tag{10}$$

providing a representation for the system output frequency response similar to the representation in the linear case.

When subject to the harmonic input (2), it can be shown that the output response of system (1) contributed by system nonlinearity at zero frequency can be represented using (10) as

$$\begin{aligned} y_{DC} &= Y(0) - y_s \\ &= 2\left(\frac{A}{2}\right)^2 G_{2\omega_F}(0) + 6\left(\frac{A}{2}\right)^4 G_{4\omega_F}(0) + \cdots \end{aligned} \tag{11}$$

In (11), $G_{2n\omega_F}(0), n=1,2,\ldots$ denotes the NOFRFs $G_{2n}(\omega), n=1,2,\ldots$ of system (1) evaluated at $\omega=0$ in the case where the system is subject to harmonic input (2) with frequency ω_F .

A comparison of Eq.(11) and Eq.(4) implies that the problems with the application of the existing GFRFs based analysis for the favorability of introducing a periodic operation could be addressed by using a NOFRFs based approach. This is because the NOFRFs can be numerically evaluated up to an arbitrary order only using the system input and output data (Lang et al., 2007).

3.2 The NOFRFs based analysis of the periodically operated chemical reaction process

The idea of the NOFRFs based analysis is to numerically determine the NOFRFs $G_{2n\omega_F}(0), n=1,2,\ldots$ in Eq.(11) using the system input and output data so as to facilitate the evaluation and optimal design of the favorable effects of a periodic operation. This can be achieved by using the method for the evaluation of the NOFRFs proposed in (Lang et al., 2007) and the system input output data which can be obtained from either model simulation or experimental test. Mathematically, this idea can be described as follows.

Assume that the component y_{DC} of the response of system (1) to harmonic input (2) under M different amplitudes is all available. Denote the M different amplitudes as

$$A_m, m=1,\ldots,M$$

and the corresponding y_{DC} as

$$y_{DCm}, m=1,\ldots,M$$

Then, it is known from Eq.(11) that

$$y_{DCm} = 2\left(\frac{A_m}{2}\right)^2 G_{2\omega_F}(0) + 6\left(\frac{A_m}{2}\right)^4 G_{4\omega_F}(0) + \cdots \quad m=1,2,\ldots,M \tag{12}$$

Consequently, the NOFRFs $G_{2n\omega_F}(0), n=1,2,\ldots$ can be determined by using a Least Squares approach as follows,

$$\begin{bmatrix} G_{2\omega_F}(0) \\ G_{4\omega_F}(0) \\ \vdots \\ \vdots \end{bmatrix} = (\bar{X}^T\bar{X})^{-1}\bar{X}^T \begin{bmatrix} y_{DC,1} \\ y_{DC,2} \\ \vdots \\ y_{DC,M} \end{bmatrix} \tag{13}$$

where

$$\bar{X} = \begin{bmatrix} 2\left(\frac{A_1}{2}\right)^2 & 6\left(\frac{A_1}{2}\right)^4 & \cdots \\ 2\left(\frac{A_2}{2}\right)^2 & 6\left(\frac{A_2}{2}\right)^4 & \cdots \\ \vdots & \vdots & \cdots \\ 2\left(\frac{A_M}{2}\right)^2 & 6\left(\frac{A_M}{2}\right)^4 & \cdots \end{bmatrix} \tag{14}$$

As $G_{2n\omega_F}(0), n=1,2,\ldots$ can be determined as described above for all relevant ω_F of interest, Eq. (11) and $G_{2n\omega_F}(0), n=1,2,\ldots$ thus determined provide a numerical relationship that can be used to analyze and design the effect of modulation parameters A and ω_F on the favorability and performance of a chemical reaction process periodic operation.

4. SIMULATION STUDY OF THE EFFECT OF MODULATION PARAMETERS ON PERIODIC OPERATION

4.1 The mathematic model of a non-isothermal CSTR

Consider a simple nonlinear homogeneous *n*-th order reaction *A→product(s)*, the rate of reaction is given by

$$r = k_0 e^{-(E_A/RT)} c_A^n \tag{15}$$

where c_A is the reactant concentration, T is the temperature, E_A is the activation energy, k_0 is the pre-exponential factor in the Arrhenius equation and R is the gas constant, respectively. The material balance and the energy balance for the reactant A can be written as

$$V\frac{dc_A}{dt} = Fc_{A,i} - Fc_A - k_0 e^{-\frac{E_A}{RT}} c_A^n V \tag{16}$$

$$V\rho c_p \frac{dT}{dt} = F\rho c_p T_i - F\rho c_p T + (-\Delta H_R) k_0 e^{-\frac{E_A}{RT}} c_A^n V - UA_W(T-T_J) \tag{17}$$

where t is the time, F is the volumetric flow rate of the reaction stream, V is the volume of the CSTR reactor, ΔH_R is the heat of reaction, A_W is the surface area of the heat exchanger, U is the overall heat transfer coefficient, ρ is the density and c_p is the specific heat capacity. Subscript i for the inlet and Subscript J for the heating/cooling fluid in the reactor jacket, respectively (Nikolić et al.,2014, 2015). All the parameters of the model equations used in the simulation are listed in Table 1 (Marlin, 2000).

Assume that Eq. (11) can, in this case, be represented as

$$y_{DC} = 2\left(\frac{A}{2}\right)^2 G_{2,\omega_F}(0) + 6\left(\frac{A}{2}\right)^4 G_{4,\omega_F}(0) \tag{18}$$

Then, the values of $G_{2,\omega_F}(0)$, $G_{4,\omega_F}(0)$ in (18) can be determined using the method in Section 3.2 from the system output data y_{DC} under the harmonic input (2) of amplitudes A_1 and A_2, respectively.

Table 1. Parameters and operating conditions of CSTR

Parameter	Value	Units
Reaction order, n	1	
Preexponential factor of the reaction rate constant, k_0	10^{10}	1/min
Activation energy, E_A	69256	kJ/kmol
Heat of reaction, ΔH_R	-543920	kJ/kmol
Heat capacity, ρCp	4.184×10^3	kJ/K/m^3
Volume of the reactor, V	1	m^3
Steady-state flow-rate, Fs	1	m^3/min
Steady-state inlet concentration, $c_{Ai,s}$	2	kmol/m^3
Steady-state inlet temperature, $T_{i,s}$	323	K
Steady-state temperature of the coolant, $T_{J,s}$	365	K
Overall heat transfer coefficient multiplied by the heat transfer area, UA_w	27337	kJ/K/min

4.2 The NOFRFs based analysis

From the input output data of system (16)(17) produced by the periodic modulation of inlet temperature, i.e.,

$$T_i(t) = T_{is} + A\cos(\omega_F t) \qquad (19)$$

in the cases of $A_1 = 10\%$, $A_2 = 20\%$ and frequency ω_F varying from 0 to 100 rad/min, the values of $G_{2,\omega_F}(0)$, $G_{4,\omega_F}(0)$ in (18) were obtained. Fig.1 shows the diagram of $G_{2,\omega_F}(0)$ as the function of log scaled frequency ω_F. Fig.2 gives the diagram of the y_{DC} as the function of log scaled frequency ω_F when the input amplitude is $A_1 = 10\%$. The obtained $G_{4,\omega_F}(0) \approx 0$ over all the frequency ranges of concern.

From Fig.1-2, it can be concluded that in the low frequency range of $\omega_F \le 5.76\,rad/\min$ and in the high frequency range of $\omega_F \ge 30\,rad/\min$, $G_{2,\omega_F}(0) > 0$ and $y_{DC} > 0$, indicating that the improvement of reactant conversion is impossible. While, when $5.76\,rad/\min < \omega_F < 30\,rad/\min$, $G_{2,\omega_F}(0) < 0$, $y_{DC} < 0$, showing that performance improvement can be expected by the introduction of periodic operation. Particularly, both $G_{2,\omega_F}(0)$ and y_{DC} have minima near $\omega_F = 7\,rad/\min$, showing, a best periodic operation performance can be reached.

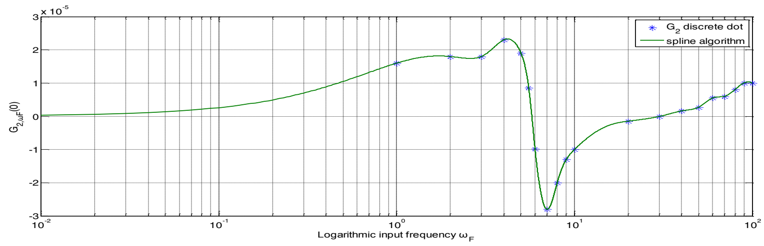

Fig.1 The NOFRF $G_{2,\omega_F}(0)$ vs. logarithmic input frequency ω_F

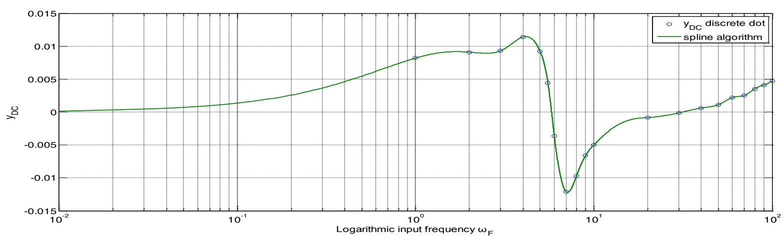

Fig.2 DC component y_{DC} when amplitude A_1=10% vs. logarithmic input frequency ω_F

Figs.3-4 shows the numerical simulation results of the outlet concentration and temperature.

It can be observed from Fig.3 that, the fluctuation ranges of output concentration and temperature are $c_A(t) \in [0.1 \;\; 0.6]\,kmol/m^3$ and $T(t) \in [370 \;\; 430]\,K$ when $A = 15\%$ and $\omega_F = 5.53\,rad/\min$. The mean value of output concentration $c_A^m = 0.3575\,kmol/m^3$ is higher than the steady state value $c_{A,s} = 0.3468\,kmol/m^3$, making the periodic operation unfavourable (as $y_{DC} = \Delta > 0$).

In Fig.4, the fluctuation ranges of output concentration and temperature are $c_A(t) \in [0.25 \;\; 0.4]\,kmol/m^3$ and $T(t) \in [380 \;\; 400]\,K$ when $A = 15\%$ and $\omega_F = 10\,rad/\min$. $c_A^m = 0.3368\,kmol/m^3$ is lower than $c_{A,s} = 0.3468\,kmol/m^3$, indicating the periodic operation is favorable. These results are obviously consistent with the NOFRFs based analysis above.

In addition, the output temperatures values in Figs.3-4 both are close to the steady state $T_s = 388K$, showing that the output temperature response is not be affected by periodic modulation of input temperatures.

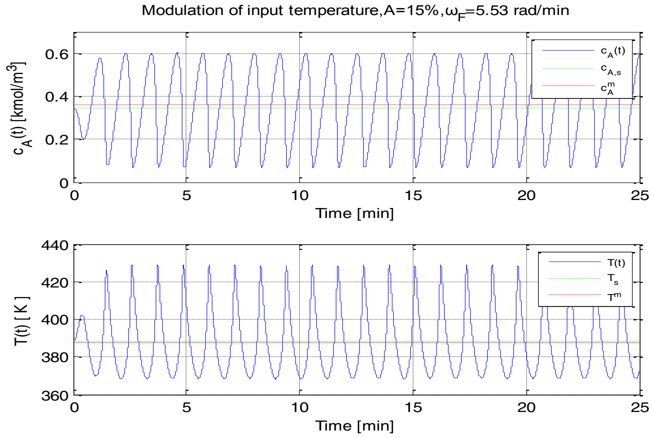

Fig.3 The output concentration and temperature when modulation of input temperature with amplitude A=15% and frequency ω_F=5.53rad/min

The values of y_{DC} calculated by numerical simulation, the existing GFRFs method, and the new NOFRFs method are compared in Table 2.

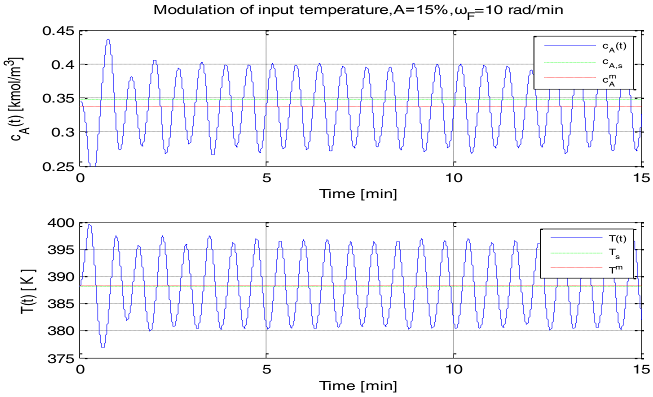

Fig.4 The output concentration and temperature when modulation of input temperature with amplitude A=15% and frequency ω_F=10 rad/min

It can be observed in Table 2 that y_{DC}, obtained by the NOFRFs based method has a better agreement with the values obtained by numerical solution in almost all the cases, demonstrating the effectiveness and advantage of the NOFRFs based new method.

Table 2. The values of y_{DC} calculated by numerical simulation, the existing GFRFs and the new NOFRFs method

	Modulation of inlet temperature											
	15% input amplitude			10% input amplitude			5% input amplitude			3% input amplitude		
Frequency ω_F	Δ_{num}	$y_{,GFRFs}$	$y_{,NOFRFs}$	Δ_{num}	$y_{,GFRFs}$	$y_{,NOFRFs}$	Δ_{num}	$y_{,GFRFs}$	$y_{,NOFRFs}$	Δ_{num}	$y_{,GFRFs}$	$y_{,NOFRFs}$
1	0.0186	0.0181	0.0185	0.0082	0.0080	0.0082	0.0020	0.0020	0.0020	0.0007	0.0007	0.0007
2	0.0192	0.0196	0.0196	0.0088	0.0087	0.0088	0.0022	0.0022	0.0022	0.0008	0.0008	0.0008
3	0.0210	0.0224	0.0208	0.0095	0.0100	0.0095	0.0025	0.0025	0.0024	0.0009	0.0009	0.0009
5	0.0200	0.0166	0.0201	0.0105	0.0074	0.0105	0.0026	0.0018	0.0028	0.0009	0.0007	0.0010
5.53	0.0119	-0.0325	0.0111	0.0053	-0.0144	0.0053	0.0003	-0.0036	0.0014	-0.0004	-0.0013	0.0005
6	0.0022	-0.0600	-0.0013	-0.0023	-0.0267	-0.0023	-0.0038	-0.0067	-0.0008	-0.0024	-0.0024	-0.0003
7	-0.0154	-0.0424	-0.0184	-0.0112	-0.0169	-0.0112	-0.0042	-0.0039	-0.0033	-0.0016	-0.0014	-0.0012
10	-0.0099	-0.0096	-0.0099	-0.0046	-0.0042	-0.0046	-0.0013	-0.0011	-0.0012	-0.0006	-0.0004	-0.0004

4.3 Optimal design using the NOFRFs

In order to achieve a minimum value for y_{DC} so as to reach a maximal conversion, it is desirable to find an optimal value for both the periodic operation amplitude A and frequency ω_F. From Tab.2, $\omega_F \in [5.53, 10] rad/\min$ is selected as the frequency boundary for the effective frequency range associated with a negative value of y_{DC}. The maximum input amplitude is determined as 15% ($\Delta T_i = 48.5K$) to ensure the temperature not to exceed 100℃. Based on the discrete data generated by the NOFRFs based analysis, a data fitted relationship between y_{DC}, A and ω_F, denoted as $y_{DC} = f(\omega_F, A)$, was obtained as follows and illustrated in Fig. 5.

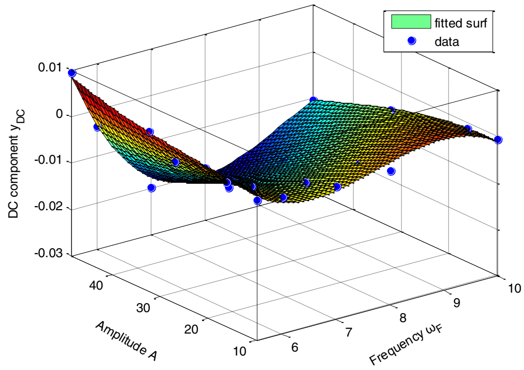

Fig.5 The data fitted surface of y_{DC}

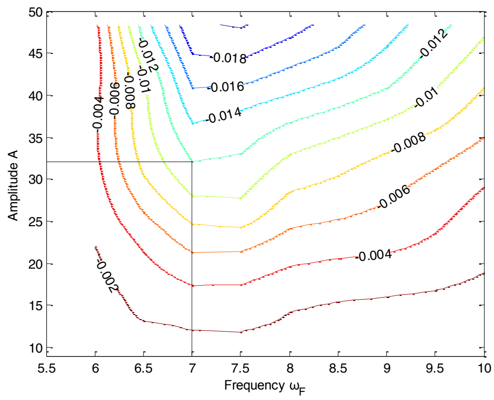

Fig.6 Contour map of the data fitted surface y_{DC}

$$y_{DC} = f(\omega_F, A) = 0.273 - 0.1187\omega_F + 0.0058A + 0.0167\omega_F^{\,2} - 0.00157\omega_F A$$
$$+3\times10^{-6}A^2 - 0.00076\omega_F^{\,3} + 0.0001\omega_F^{\,2}A - 2\times10^{-6}\omega_F A^2 + 1\times10^{-7}A^3 \quad (20)$$

Fig.5 shows that y_{DC} has a global minimum within the effective boundaries of amplitude A and frequency ω_F, which is the solution to the following optimization problem,

$$\min f(\omega_F, A)$$
$$s.t. \quad 5.53\,rad/\min \le \omega_F \le 10\,rad/\min; \quad (21)$$
$$9K \le A \le 48.45K$$

and can be obtained as

$$\omega_F = 7.8\,rad/\min$$
$$A = 48.45K \text{ (15\% input amplitude)}$$

producing

$$y_{DC,\min} = -0.02\,kmol/m^3$$

The contour map of the surface $y_{DC} = f(\omega_F, A)$ is shown in Fig. 6 which is the relationship between amplitude A and frequency ω_F for a given y_{DC}. For example, given $A = 32K$ (10% amplitude), the input frequency ω_F can be found to be $7\,rad/\min$ if an optimal conversion with $y_{DC} = -0.012\,kmol/m^3$ is to be reached. These demonstrate the significance of the NOFRFs based design for a desired periodic operation.

5. CONCLUSIONS

In the present study, a new NOFRFs based approach is proposed to analyse and design the periodic operation of a non-isothermal CSTR with a periodically modulated inlet temperature. A comparison between the existing GFRFs based method and the new NOFRFs approach has demonstrated the effectiveness and advantage of the new method. In addition, an optimal design of the periodic operation parameters using the NOFRFs approach has been conducted. The results have shown that the NOFRFs approach has potential to perform an optimal design to reach a maximum conversion of the reactant, which cannot be achieved by existing techniques.

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