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Nonlinearity assessment of chemical processes

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Abstract. One of the most important issues in controller design and analysis of a nonlinear system is its degree of nonlinearity. Helbig et al. proposed a method in which the system, along with a linear reference model (which is the sum of a couple of first order transfer functions), is stimulated with a set of input patterns. The outputs of these systems are then compared and the nonlinearity measure of the system is obtained as the normalized difference between the outputs of these two systems. In this paper, the linear reference model is replaced with two simpler linear ones. The proposed method has been used for assessment of the nonlinearity measure of various nonlinear systems that are used as standard benchmarks by the nonlinear process control research community. The results show that, despite the use of this simpler model with less parameters, the calculated nonlinearity measure is almost similar to the one obtained by Helbig's method. Furthermore, the nonlinearity measure obtained by the proposed method can be calculated very much faster.

1. Introduction

Measurement of process nonlinearity in chemical processes is the most important part of the identification and description of a system for the design of a suitable control structure. This is because of the inherent nonlinear behavior of all chemical processes. Nonlinearity of a system can be defined as the measure of nonlinearity in the input-output behavior of the system. Analysis of a nonlinearity measure can be classified into open-loop and closed-loop analyses: Investigation of open-loop systems only leads to identification of the behavior of the process, whereas, studying closed-loop systems further indicates the effect of nonlinearity in assessment of closed-loop control systems. In addition, this is useful in the design of optimal controllers [1]. The criteria used in the Non-Linearity Measure (NLM) is the difference between the best adapted parallel linear system and the real nonlinear process for the various and worst different inputs [2,3]. One can use the NLM to judiciously select the appropriate algorithm for identification and then control of the process. Over the last three decades, a variety of nonlinear controller design techniques have been developed which need constraints and easy low order models, whilst the main model of the process is more complex. For instance, a distillation column contains a large number of algebraic and differential equations, which makes these control algorithms either impossible or extremely difficult to use for distillation column control. Another instance of such a case would be the control of blood glucose levels in patients suffering from diabetes, based on the detailed physiological models proposed in the literature [4,5]. In this study, the measure of nonlinearity proposed by Helbig et al. [2] is extended, such that it may be obtained faster and with less computational demands.

A variety of linear systems are modified in the literature in order to approximate nonlinear systems for

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model based nonlinearity assessments; A brief review is presented in [6]. Helbig et al. [2] proposed a weighted summation of several first order transfer functions with various time constants for this purpose.

In this work, Helbig's method has been extended, both in speed of assessment and its generality, to be able to address the processes with time delays, various types of oscillations and by reducing its sensitivity to the parameters of linear models. A Second Order Plus Dead-Time (SOPDT) linear model, along with a zero order model, has been added to the set of candidate linear models used to assess system nonlinearity measures, rather than the weighted summation of several first order systems. This extension makes it possible to assess the NLM in a fraction of a second, which is noticeably less than the required time in the others. It would be significantly helpful in Real-Time Optimization (RTO) and plant-wide control of the process plants, which have a nonlinear dynamic under a wide range of operating conditions (which are dictated by the RTO in the Supervisory Control Layer) in which the validity of the linear model has to be assessed.

The problem is presented in the form of a convex optimization problem [2], and, finally, the numerical convex optimization methods are used to calculate the NLM of the system. In order to show the performance of the extended scheme and its improvements for systems with various types of nonlinearity, it has been used to assess the NLM of several nonlinear chemical processes. The following seven standard benchmarks are used in nonlinear system analysis and control articles: (1) a mildly nonlinear CSTR, (2) the Van de Vusse process, (3) a fermentation reactor, (4) a pH neutralization system with and without measurement delay, (5) a high purity distillation column, (6) two nonlinear CSTRs in series, and (7) the Sorensen model of Type I Diabetes Mellitus (T1DM) disease. Simulations of the aforementioned processes are performed according to their state space models, except for the fifth benchmark, which has been simulated by a commercial dynamic simulator. Various input patterns have been used in the stimulation of the models, and the outputs of simulations are saved for each input pattern.

The paper is organized as follows. A brief literature review is presented in Section 2. In Section 3 the NLM is defined and formulated in the form of an *inf-sup-inf*-problem whose optimum point is not feasible. The resulted problem is transformed to a finite convex optimization problem on the space of linear time-delayed systems in Sections 4. Afterwards, in Section 5, the performance of the proposed method has been shown via its application for assessment of the NLM of several nonlinear chemical processes with various types of nonlinearity. The paper is concluded in Section 6.

2. Literature review

An overall view of nonlinearity measures in the literature can be acquired through [6-8]. The goal is to realize how severe the degree of nonlinearity of the process is and to see if linear control strategies can be used to control the system.

The Regression Error Specification Test, which is a popular linearity test method and does not depend on any assumption, is introduced by Ramsey [9]. Tan [6] classified this method in the class of a statistical approach, as well as [10-14].

Desoer & Wang [15] proposed the NLM in the form of a *min*-problem that quantifies the difference between the dynamical outputs of a nonlinear system, N, and a parallel well defined linear system, G:

$$\phi_N^{\mathcal{U}} = \inf_{G\in\mathcal{G}} \sup_{u\in\mathcal{U}} \|N[u] - G[u]\|, \tag{1}$$

where $\mathcal{U}$ and $\mathcal{G}$ denote the set of input signals and the set of admissible linear systems, respectively, and operator $\|.\|$ is the induced norm. They also employed the equation:

$$\phi_N^{\mathcal{U}} = \inf_{G\in\mathcal{G}} \sup_{u\in\mathcal{U}} \frac{\|N[u] - G[u]\|}{\|u\|}, \tag{2}$$

that is a normalized form of Eq. (1) with the induced norm of input, u.

Also, Nikolaou [16] introduced a notation of the inner product in order to quantify the nonlinearity of a dynamic system. The measure can be computed by considering an appropriate input set and calculation of the NLM using Eq. (1) by Monte-Carlo simulations.

Ogunnaike et al. [17] defined the NLM as the maximum difference between any pair of locally approximating linear systems around the operating region.

Stack & Doyle III [18,1] proposed a control relevant NLM in the sense of coherence analysis. They used the nonlinear system as a stationary random process and proposed the input-output coherence as the NLM of the system.

Allgöwer [19] introduced the NLM in the form of:

$$\phi_N^{\mathcal{U}} = \inf_{G\in\mathcal{G}} \sup_{u\in\mathcal{U}} \frac{\|N[u] - G[u]\|}{\|N[u]\|}, \tag{3}$$

which is the normalized Eq. (1), with the induced norm of nonlinear output, N, rather than input in Eq. (2).

Alternatively, Helbig et al. [2] modified the notion of Eq. (3) by considering initial conditions of the nonlinear system in addition to input space in Eqs. (1)-(3):

$$\phi_N^{\mathcal{U}} = \inf_{G\in\mathcal{G}} \sup_{(u,x_{N,0})\in\mathcal{S}} \inf_{x_{G,0}\in\mathcal{X}_{G,0}} \frac{\|N[u, x_{N,0}] - G[u, x_{G,0}]\|}{\|N[u, x_{N,0}]\|}, \tag{4}$$

where $\mathcal{S} = \{(u, x_{N,0}) : u \in \mathcal{U}, x_{N,0} \in \mathcal{X}_{N,0}\}$, and $\mathcal{X}_{G,0}$ and $\mathcal{X}_{N,0}$ denotes the appropriate initial conditions of the approximate linear and nonlinear systems, respectively. The motivation is to generalize Eq. (3) to make it possible to assess the nonlinearity measure for time varying processes.

Schweickhardt [8] used minimal linear models to compute the NLM based on Eq. (3). According to this method, an appropriate structure for the linear model is chosen and then the minimal modeling error of the nonlinear and best linear system is used as NLM.

Sun & Hoo [20] proposed a NLM for SISO nonlinear systems by the following equation:

$$\phi_N^{\mathcal{U}} = \max\{\sup_{u\in\mathcal{U}} \|N[u] - G_{up}[u]\|, \sup_{u\in\mathcal{U}} \|N[u] - G_{lo}[u]\|\}, \quad (5)$$

where G_{up} and G_{lo} are linear systems, such that the output of the nonlinear system bounds at all times between the outputs of the linear systems, G_{up} and G_{lo}.

A Gramian based NLM approach was presented by Hahn & Edgar [21]. Empirical controllability and observability Gramians are used in this notion to compute input-output NLM using input-state NLM and state-output NLM, respectively. The required Gramians are obtained from the nonlinear process data and are compared to Gramians from a linear system to get NLM.

The NLM computing is based on differential geometry proposed by Guay [22], which calculates the measure of closed-loop nonlinearity rather than open-loop measures previously mentioned. The aforementioned procedure uses the curvature of the steady state map as NLM extended in [23,24] to implement dynamic systems using *Frećhet* derivative operators. The dynamical approach seems to be complicated and significant simplifications should be considered, otherwise it would be impractical.

Harris et al. [25] measured system nonlinearity via the method proposed by Allgöwer [19] using functional expansions. This approach is computationally inexpensive and derives approximate lower and upper bounds on the NLM of the process.

Tan [6] proposed a NLM based on a topological approach to quantify closed-loop nonlinearities. The quantification of the distance between the nonlinear system under study and its linearized model in a closed-loop mode is given by the use of a ν-gap metric. Finally the largest ν-gap, which is a result of closed-loop nonlinearity, can be obtained. Hosseini [26] modified an H-gap metric based method and used it in model bank selection in multi-linear model analysis. Also, Du et al. [27] and Du & Tong [28] used a gap metric based method for NLM calculation. The authors tried to simplify the calculations and reduce the computation time by smart selection of gridding points. However, they mentioned that the current gap metric based approaches are computationally intensive.

Li [29] proposed a general measure of nonlinearity for stochastic systems, which is based on the quantification of the deviation of the system from linearity.

In this paper, the NLM is investigated by an extended approach, based on the proposed method in [2]. It seems to be more general and applicable for a larger set of systems compared to its alternatives.

3. Definition of nonlinearity measurement

The NLM of a process is defined as the normalized difference between nonlinear and parallel linear systems. Investigation of the dynamic behavior of a nonlinear system starts by choosing the manipulating variables and the outputs of the system. The inputs should be those which are used as manipulating variables to control the selected outputs. The required data to calculate the NLM are the time series of the selected pairs, which can be obtained from the real plant or its mathematical models. Indeed, the only important data are the required input-output data obtained from the system itself (if it has not been modeled yet) or its simulation, based on the mathematical model representing the system, whose inputs and outputs are $u_{N,in}(t)$ and $y_{N,out}(t)$, respectively. This system would be in the form of the state space of Eq. (6):

$$\dot{\boldsymbol{x}}_N = \boldsymbol{f}(\boldsymbol{x}_N,\ \boldsymbol{u}_{N,in}),\quad \boldsymbol{x}_N(0) = \boldsymbol{x}_{N,0}, \quad (6)$$

$$\boldsymbol{y}_{N,out} = \boldsymbol{h}(\boldsymbol{x}_N,\ \boldsymbol{u}_{N,in}).$$

In Eq. (6), the inputs of the nonlinear process ($\boldsymbol{u}_{N,in}$), the states ($\boldsymbol{x}_N$) and the outputs ($\boldsymbol{y}_{N,out}$) are, respectively, members of spaces $\mathcal{U}_{in}$, $\mathcal{X}$, and $\mathcal{Y}_{out}$. The space, $\mathcal{U}_{in}$, contains the total applicable inputs of the specified process, and the spaces, $\mathcal{X}$ and $\mathcal{Y}_{out}$, represent the states and outputs of the system, respectively.

Deviation variables are defined to implement calculations in the general form. Here, the deviation variables are defined, with respect to reference values, as $\boldsymbol{u}_N = \boldsymbol{u}_{N,in} - \boldsymbol{u}_{N,s}$ and $\boldsymbol{y}_N = \boldsymbol{y}_{N,out} - \boldsymbol{y}_{N,s}$. u_N and y_N are members of $\mathcal{Y}_N = \{\boldsymbol{y}_N : (\boldsymbol{y}_N + \boldsymbol{y}_{N,s}) \in \mathcal{Y}_{out}\}$ and $\mathcal{U} = \{\boldsymbol{u}_N : (\boldsymbol{u}_N + \boldsymbol{u}_{N,s}) \in \mathcal{U}_{in}\}$, respectively. The reference values for continuous processes are the values of variables at the operating point. By applying the new deviation variables, the described system in Eq. (6) is rewritten as Eq. (7). The related equation of the approximated linear system is defined the same as a nonlinear system.

$$\boldsymbol{y}_N = N[\boldsymbol{u}_N, \boldsymbol{x}_{N,0}], \quad (7)$$

$$\boldsymbol{y}_G = G[\boldsymbol{u}_G, \boldsymbol{x}_{G,0}]. \quad (8)$$

The nonlinearity measure of the dynamic nonlinear system, $N : \mathcal{U} \times \mathcal{X}_{N,0} \rightarrow \mathcal{Y}$, with output y_N, is defined as:

$$\phi_N^{\mathcal{U}} = \inf_{G \in \mathcal{G}} \sup_{(\boldsymbol{u}, \boldsymbol{x}_{N,0}) \in \mathcal{S}} \inf_{\boldsymbol{x}_{G,0} \in \mathcal{X}_{G,0}} \frac{\|N[\boldsymbol{u}, \boldsymbol{x}_{N,0}] - G[\boldsymbol{u}\ , \boldsymbol{x}_{G,0}]\|}{\|N[\boldsymbol{u}, \boldsymbol{x}_{N,0}]\|}, \tag{9}$$

where $G : \mathcal{U} \times \mathcal{X}_{G,0} \rightarrow \mathcal{Y}$ is a member of space $\mathcal{Y}$ and $\mathcal{S} = \{(u, x_{N,0}) : u \in \mathcal{U}, x_{N,0} \in \mathcal{X}_{N,0}\}$. In Eq. (9), $\phi_N^{\mathcal{U}}$ is the maximum normalized difference between the nonlinear system, N, and the parallel linear system, G, corresponding to the worst input signals and initial conditions of the nonlinear system, N, and the best initial conditions of the parallel linear system, G. The first *inf*-operator optimizes the linear system, G, in space $\mathcal{G}$, in order to approximate N as closely as possible. The *sup*-operator creates the worst conditions of the nonlinear system by the worst inputs and initial conditions. The second *inf*-operator finds the best initial conditions for the parallel linear system, G.

The space, $\mathcal{S}$, should be considered so that the corresponding outputs of the system remain bounded, otherwise the calculations will encounter several problems. The suitable and appropriate choice of inputs is an important part of the algorithm. The input signals should be chosen such that the considered operating range can be covered as much as possible; furthermore in closed-loop systems, the admissible signal should comprise the stability of the closed-loop. It should be noted that the sampling time and time span $[t_0, t_f)$ of the operation are consequential as well.

The nonlinearity measure, $\phi_N^{\mathcal{U}}$, is a positive real number between zero and one. The value of zero for NLM, $\phi_N^{\mathcal{U}} = 0$, means that the approximated parallel linear system exactly coincides with the system, and the considered process is exactly linear. On the other hand, $\phi_N^{\mathcal{U}} = 1$ implies that the system has quite a nonlinear behavior and the parallel linear system is not coincident with the nonlinear system. The fundamental characteristics of the HLM and their corresponding theoretical basis have been discussed by Sun & Hoo [20].

4. Computation of nonlinearity measure

According to the proposed equation by Helbig et al. [2] in Eq. (4), calculation of NLM requires the solution of an *inf-sup-inf*-problem, whose corresponding optimum point is not feasible. As a solution to convert the problem to a *min-max-min*-problem, spaces $\mathcal{G}$, $\mathcal{S}$ and $\mathcal{X}_{G,0}$ should be restricted in an acceptable interval.

A suitable linear set, $\mathcal{G}_c$, which is dense in $\mathcal{G}$, should be considered to constrain space $\mathcal{G}$. Different sets can be used to accomplish this task. Space $\mathcal{S}_c$ is an approximation of space $\mathcal{S}$ with n members. As a result, the *sup*-operator is transformed to a *max*-operator. After enforcing the changes, a *min-max*-problem is achieved. Finally, a constrained *min* optimization problem is achieved by using a virtual variable, z, as shown in Eq. (10) for all $(u_j, \boldsymbol{x}_{N,0_j}) \in \mathcal{S}_c$. The solution of this optimization problem ($\theta_N^{\mathcal{S}_c}$) is an approximation of $\phi_N^{\mathcal{U}}$. In fact, the target function is converted to a real number, $z \in \Re$, and the eliminated *min*- and *max*-operators are observed in the added constraints.

$$\theta_N^{\mathcal{S}_c} = \min_{z \in \Re, G \in \mathcal{G}_c} z,$$
$$\text{s.t.} \frac{\|N[u_j, \boldsymbol{x}_{N,0_j}] - G[u_j, \boldsymbol{x}_{G,0_j}]\|}{\|N[u_j, \boldsymbol{x}_{N,0_j}]\|} - z \leq 0. \tag{10}$$

In [2], to simplify the problem, another reasonable assumption is considered. Since, for a continuous stable process to reach its specific equilibrium point, providing it works at sufficiently fast large amount of time regardless of its initial condition, the internal *inf*-operator can be disregarded and the initial values can be set to zero for all linear systems. Thus, Eq. (4) is rewritten as Eq. (11):

$$\phi_{NOP}^{\mathcal{U}} = \inf_{G \in \mathcal{G}} \sup_{u \in \mathcal{U}} \frac{\|N[\boldsymbol{u}, \boldsymbol{x}_{N,s}] - G[\boldsymbol{u}, \boldsymbol{0}]\|}{\|N[\boldsymbol{u}, \boldsymbol{x}_{N,s}]\|}. \tag{11}$$

Hence, after implementation of the aforementioned assumptions, the problem is transformed into a finite convex optimization problem:

$$\theta_{NOP}^{\mathcal{S}_c} = \min_{z \in \Re, G \in \mathcal{G}_c} z,$$
$$\text{s.t.} \frac{\|N[u_j, \boldsymbol{x}_{N,s_j}] - G[u_j, \boldsymbol{0}]\|}{\|N[u_j, \boldsymbol{x}_{N,s_j}]\|} - z \leq 0. \tag{12}$$

To calculate $\theta_{NOP}^{\mathcal{S}_c}$ using Eq. (12), spaces $\mathcal{G}_c$ and $\mathcal{U}_c$ are required to be properly specified. The set, $\mathcal{U}_c$, is a collection of inputs which are applied to the nonlinear system and parallel linear system as well. This collection should be created in such a way that the outputs be bounded and reasonable. This input collection can consist of any kind of input; step, pulse, ideal pulse, random number and others. The kind of input has no effect on the results [2,3]. The resulted convex optimization problem could be efficiently solved by various optimization algorithms proposed for this type of problem. In this study, an interior-point algorithm [30] has been used to solve the obtained convex optimization problem.

To approximate the behavior of the nonlinear system, three sets of transfer function are used separately: (1) the weighted sum of the first order linear transfer functions, (2) a Second Order transfer function

Plus Dead Time (SOPDT), and (3) a zero order (ZO) transfer function. The SOPDT and ZO systems are introduced here, while the first system is proposed by Helbig et al. [2]. The linear systems are illustrated in Eqs. (13)-(15), respectively; moreover, a direct feedthrough is added to each system.

$$G_{FO}[u, \boldsymbol{x}_{G,0}] = \sum_{i=1}^{m} d_i.G_i[u, \boldsymbol{x}_{G,0_i}] + d_{m+1}.u,$$

$$G_i(s) = \frac{1}{1+T_i s}, \boldsymbol{T} \in \Re_+^m, \tag{13}$$

$$G_{SO}[u, \boldsymbol{x}_{G,0}] = \frac{K.e^{-Ds}}{\tau^2 s^2 + 2\tau\xi s + 1} + d.u, \tag{14}$$

$$G_{ZO}[u, \boldsymbol{x}_{G,0}] = K.u. \tag{15}$$

The first is proposed and utilized in [2]. There are $m+2+m.n$ decision variables in Eq. (13), which are considerable in comparison to second order and zero order transfer functions. This is the cause of a huge reduction in computation time. The other advantage of SOPDT used as a parallel linear system is to make the method applicable for nonlinear systems with time delay, as illustrated in an example of pH neutralization in section 5.5.

If it is assumed that the operating point of the system is transformed to zero by the use of deviation variables, Eqs. (13)-(15) are rewritten as Eqs. (16)-(18):

$$G_{FO}[u, \boldsymbol{0}] = \sum_{i=1}^{m} d_i.G_i[u, \boldsymbol{0}] + d_{m+1}.u,$$

$$G_i(s) = \frac{1}{1+T_i s}, \boldsymbol{T} \in \Re_+^m, \tag{16}$$

$$G_{SO}[u, \boldsymbol{0}] = \frac{K.e^{-Ds}}{\tau^2 s^2 + 2\tau\xi s + 1} + d.u, \tag{17}$$

$$G_{ZO}[u, \boldsymbol{0}] = K.u. \tag{18}$$

By this assumption, Eq. (13) is simplified and the number of decision variables in the optimization problem is considerably reduced. The number of variables in Eq. (13) is $m+2+m.n$, where $m+1$ represents the number of d's, one variable designates z, and $m.n$ variables correspond to the initial conditions of the linear first order transfer functions. When the operating point is shifted to zero, the corresponding variable in the problem is eliminated. Therefore, the number of decision variables is reduced to $m+2$ instead of $m+2+m.n$ variables. The suggested second and zero order transfer functions in Eqs. (17) and (18) have 6 and 2 decision variables, respectively. Hence, in the established framework, the number of decision variables is a lot less than those of the original framework.

5. Case studies

The problem is presented in the form of a convex optimization problem. The performance of the introduced scheme is investigated in seven benchmark nonlinear chemical processes, with various types of nonlinearity from mildly to highly nonlinear systems: (1) mildly nonlinear CSTR [31], (2) the Van de Vusse process [32], which is highly nonlinear, (3) a fermentation reactor [33] with a moderate nonlinearity, (4) a pH neutralization system with and without measurement delay [34]; the pH system is a moderate to high nonlinear system, (5) the highly nonlinear high purity distillation column [35] which is studied like a commercial plant, (6) two nonlinear CSTRs in series [36] to show the effect of relative order of the system; this is a moderately nonlinear system, and (7) the Sorensen model of Type I Diabetes Mellitus (T1DM) disease [4]; the model of this process has numerous states and one output.

The problems are solved and the results are reported, as well as the corresponding computation times (with core i5-2450M, 4GB memory, Windows 7 OS).

5.1. Example 1

In this example, a CSTR that has mildly nonlinear behavior is considered and the performance accuracy of the introduced linear systems is compared to the proposed models of [2]. The equations representing the mathematical model of the process are represented by Pottmann & Seborg [31]:

$$\frac{dc_A}{dt} = \frac{q}{V}(c_{Af} - c_A) - k_0 c_A \exp(-\frac{E}{RT}), \tag{19}$$

$$\frac{dT}{dt} = \frac{q}{V}(T_f - T) + \frac{(-\Delta H)k_0 c_A}{\rho C_p}\exp(-\frac{E}{RT}) + \frac{\rho_c C_{pc}}{\rho C_p V} q_c[1 - \exp(-\frac{hA}{q_c \rho_c C_{pc}})](T_c - T). \tag{20}$$

The model parameters in Eqs. (19) and (20) are adapted from [31]. The initial conditions are $c_{A,0} = 8.823 \times 10^{-2}\ \frac{\text{mol}}{\text{L}}$ and $T_0 = 441.22$ K. The manipulating input is the flow rate of cooling water $u_{N,in} = q_c$, and $u_{N,s} = 100\ \frac{\text{L}}{\text{min}}$. The input region is $|u_N| = 10\ \frac{\text{L}}{\text{min}}$ in the vicinity of the steady state operating point. The time interval of simulation is $[0, 20)$ min, with a sampling time of $\Delta t = 0.1$ min. The three mentioned parallel linear systems are investigated separately. The parameters are optimized and the result is reported. The optimization problem is solved and the measure of nonlinearity, $\theta_{N_{OP}}^{\mathcal{S}_c}$, is calculated according to the mentioned input variables for the first state (c_A).

5.1.1. Weighted summation of several first order transfer functions (G_{FO})

The weighted sum of several first order transfer functions illustrated in Eqs. (13) and (16) is used as a parallel linear system. In this study, 20 first order transfer functions and a direct feedthruogh have been used as the parallel linear model. The time constants of the transfer functions are considered in the span of $2\pi \times 10^{-3}$ and $2\pi \times 10^{3}$ min with a logarithmically equal distribution. The applied inputs to the system are 14 steps at time $t = 0$. Figure 1 shows the step response of the first state (c_A) at the desired operating point.

The optimization problem is solved under the aforementioned solving conditions and the nonlinearity measure has been obtained as $\theta^{\mathcal{U}_c}_{NOP} = 0.2179$. The aforementioned optimized linear and approximated system outputs are indicated in Figure 2(a). The computation time is nearly 388 sec.

In [2], the NLM is calculated as $\theta^{\mathcal{U}_c}_{NOP} = 0.248$, based on a simulation for the time interval of $[0, 70)$ min, and the collection, $\mathcal{U}_c$, contains 140 different inputs in which other conditions are identical [2]. The above 140 inputs contain 8 step inputs and 132 random inputs, which change every 0.1 min. However, this value has been reported as $\theta^{\mathcal{U}_c}_{NOP} = 0.244$ in [37]. Additionally, the NLM of the input-output pair of $q_c - c_A$ has been obtained as $\theta^{\mathcal{S}_c}_{N} = 0.215$, with the use of free initial conditions for the transfer function, G_{FO} [2]. The number of decision variables in the original method is $140 \times 20 + 22 = 2822$. However, using deviation variables reduces the number of decision variables to $21 + 1 = 22$. Therefore, the computational demand and time of the simplified framework are much less than those of the accurate one, while there is no significant difference between the calculated NLM.

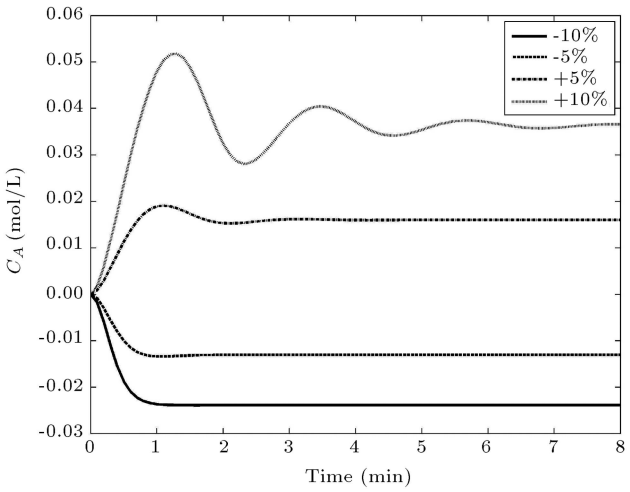

Figure 1. Transient behavior of c_A.

Figure 2(a) shows the best outputs of the linear system, which is achieved to approximate the nonlinear input-output pair, $q_c - c_A$. As seen in Figure 1, the system under study is slightly nonlinear and the outputs of the optimized parallel linear system (Figure 2(a)) exhibited an accurate approximation of the outputs. According to definition, due to the difference between the nonlinear system under examination and the optimized parallel linear system, the little difference between these parameters indicates the slight nonlinearity of the systems.

5.1.2. A second order transfer function (G_{SO})

In this part, the linear system is considered to form G_{SO} in the form of Eq. (17). This system, without considering dead time, has 5 optimized variables in the optimization problem, which, in comparison with 22 and 2822 variables in [2], reduces the required time to solve the problem. The value of nonlinearity measures is calculated as $\theta^{\mathcal{U}_c}_{NOP} = 0.2334$ for a pair of $q_c - c_A$, and the computation time is 63.04 sec. The optimized linear system's outputs are indicated in Figure 2(b).

5.1.3. A zero order transfer function (G_{ZO})

In this section, the linear system is considered to form $G_{ZO}[u, 0] = K.u$. This system has one optimized variable in the optimization problem. The optimized problem has two variables, with respect to z as the virtual variable. The value of the nonlinearity measures is calculated as $\theta^{\mathcal{U}_c}_{NOP} = 0.2526$ for the pair of

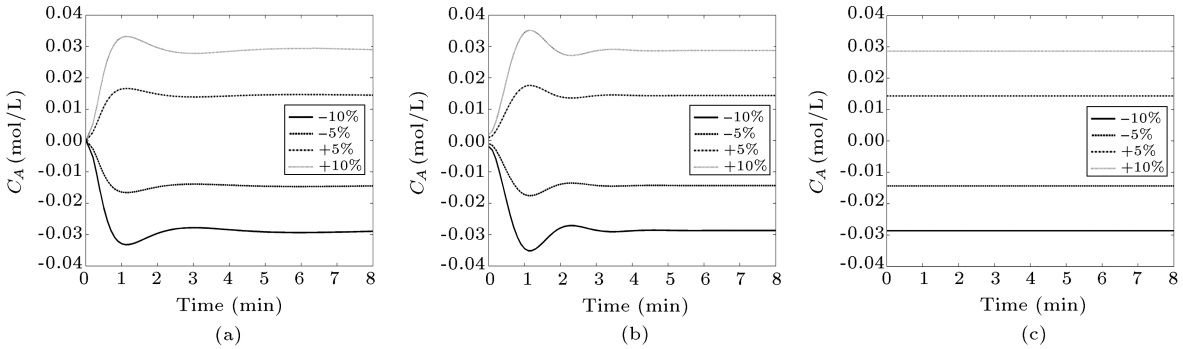

Figure 2. Changes of c_A for 4 different steps imposed on the approximated linear models for i-o pair of $q_c - c_A$: a) G_{FO}; b) G_{SO}; and c) G_{ZO}.

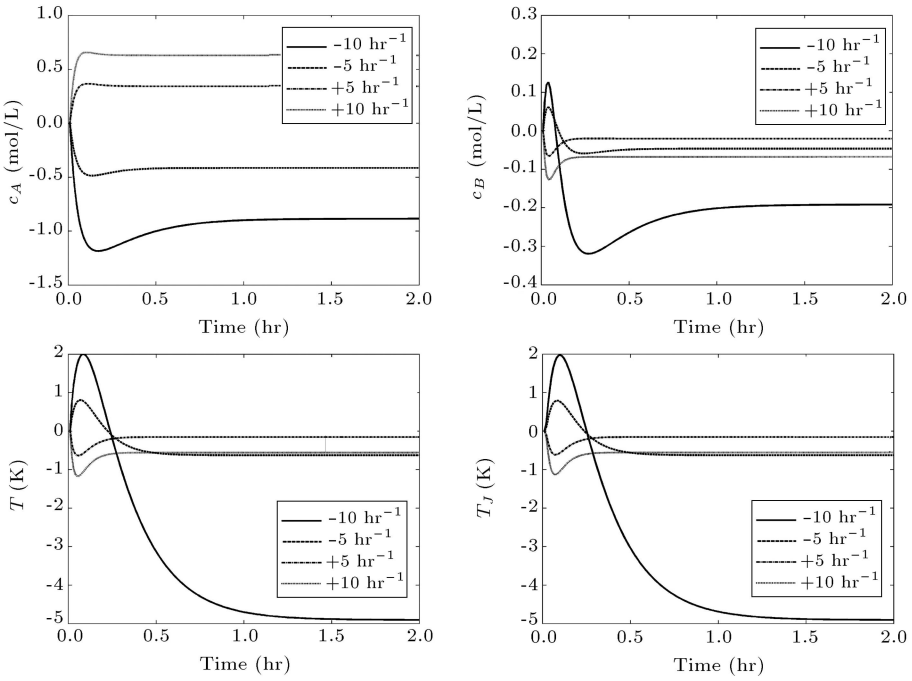

Figure 3. Changes of states in Van de Vusse reaction for 4 different inputs.

Table 1. The calculated NLM with different linear approximations in Example 1.

Linear system	$\theta^{\mathcal{U}_c}_{N_{OP}}$	t_{cal} (sec)
G_{FO}	0.2179	388.3649
G_{SO}	0.2334	63.0432
G_{ZO}	0.2526	0.1053
$G_{FO[2]}$	0.248	-
$G_{FO,[2],x_0}$	0.215	-

$q_c - c_A$, with computation time $t_{cal} = 0.1053$ sec. The optimized linear system outputs are indicated in Figure 2(c), besides the response of previous ones, G_{FO} and G_{SO}.

Additionally, in order to consider unseen operating points, in the case of continuous chemical processes like this one and also the other examples, a bias term can be added to the linear model of Eq. (18):

$$G_{ZO}[u, y_0] = b + K.u. \tag{21}$$

Figure 2 shows that the results of linear system optimization of the first (G_{FO}) and second order transfer functions (G_{SO}) are nearly the same.

The results in Table 1 show a good performance of G_{SO} according to the results of G_{FO}. Again, the estimation of NLM utilizing the very simple linear system, G_{ZO}, gives almost similar outcomes to the results of G_{SO} and G_{FO} with very small computational cost. NLM estimation employing G_{ZO} can be used, for example, in online applications that require a fast estimation of NLM.

5.2. Example 2

In this section, the nonlinearity measure of the Van de Vusse process states, along with its cooling system, are studied with respect to system inputs.

$$A \xrightarrow{k_1} B \xrightarrow{k_2} C, \tag{22}$$

$$2A \xrightarrow{k_3} D. \tag{23}$$

The CSTR processing the Van de Vusse reaction is a benchmark system for nonlinear controller design and is investigated in many research articles. In the investigated operating point, the behavior of this system is highly nonlinear and the gain of the system is changing, as illustrated in Figure 3. The states of this system are a concentration of substance A (c_A), a concentration of substance B (c_B), reactor temperature (T), and the temperature of cooling water in jacket (T_J) which are illustrated in Eqs. (24)-(27), respectively.

$$\frac{dc_A}{dt} = \frac{F}{V_R}(c_{A0} - c_A) - k_1 c_A - k_3 c_A^2, \tag{24}$$

$$\frac{dc_B}{dt} = -\frac{F}{V_R}c_B + k_1 c_A - k_2 c_B, \tag{25}$$

$$\frac{dT}{dt} = \frac{F}{V_R}(T_0 - T) + \frac{k_w A_R}{\rho C_p V_R}(T_J - T) - \frac{1}{\rho C_p}\big(k_1 c_A \Delta H_{r,AB} + k_2 c_B \Delta H_{r,BC} + k_3 c_A^2 \Delta H_{r,AD}\big), \tag{26}$$

$$\frac{dT_J}{dt} = \frac{1}{m_J C_{p,J}} \left(dotQ_J + k_w A_R (T - T_J) \right), \qquad (27)$$

$$k_i = k_{i,0} \exp(-\frac{E_i}{RT}), \quad i = 1, 2, 3.$$

The parameters of Eqs. (24)-(27) are replaced from [32]. The values of the steady state of the system are $c_{A,s} = 2.1402 \ \frac{\text{mol}}{\text{L}}$, $c_{B,s} = 1.0903 \ \frac{\text{mol}}{\text{L}}$, $T_s = 387.3411 \ K$, and $T_{J,s} = 386.0566 \ K$. The manipulating variable in this process is $u_{N,in} = \frac{F}{V_R}$ and its steady state value is $u_{N,s} = \frac{141.9}{10} = 14.19 \ \text{hr}^{-1}$. The set of $\mathcal{U}_c$ contains 20 step inputs in an interval of $-11.19 \ \text{hr}^{-1} \leq u_N \leq 11.19 \ \text{hr}^{-1}$. (This range corresponds to the deviation of the input from its absolute value.) Figure 3 shows the dynamic behavior of the nonlinear system stimulated by 4 different step inputs, which are members of $\mathcal{U}_c$. The process is simulated in the time span of $[0, 2)$ hr with sampling time $\Delta t = 0.01$ hr.

Figure 3 shows that all states have a sign change in the steady state gain except for the concentration of A, leading to $\theta^{\mathcal{U}_c}_{NOP} = 1$ [38]. Similar to Example 1, the NLM is computed for all states of the process and compared to those values calculated in the literature. Computation is accomplished using three previously discussed linear systems.

5.2.1. Weighted summation of several first order transfer functions (G_{FO})

The weighted sum of first order transfer functions in the form of Eq. (16), with 20 first order transfer functions, is used. The time constant of the transfer function is considered in the span of $2\pi \times 10^{-3}$ to 2π hr with a logarithmic equal distribution. The results of computations are presented in Table 2. The value of $\theta^{\mathcal{U}_c}_{NOP} = 0.9883$ is calculated for the input-output pair $u_N - c_B$ by the time of computation, $t_{cal} = 431.8$ sec. This number is reported as 1 and 0.9912, respectively in [2,38]. Also the values of $\theta^{\mathcal{U}_c}_{NOP}$ are reported as 0.2478, 0.9921 and 0.9957 for c_A, T and T_J, respectively [38].

5.2.2. A second order transfer function (G_{SO})

As mentioned before, the optimization problem has 5 variables. The optimization problem is solved and the results are shown in Table 2.

5.2.3. A zero order transfer function (G_{ZO})

The parallel linear system is considered in the form of $G_{ZO}[u, 0] = K.u$. The optimization problem has two variables. The optimization problem is solved and the results are recorded in Table 2.

As seen in Table 2, the results of NLM computation by the proposed linear systems are significantly acceptable in comparison to the reported results of the original framework used in [2]. Hereupon, in the rest of the examples, the SOPDT transfer function (G_{SO}) is used to assess the NLM of the benchmark nonlinear systems, and show its capabilities.

5.3. Example 3

In this example, a continuous fermentation system is studied. The nonlinear behavior of continuous fermenters is one of the interesting case study examples in nonlinear control [33]. This system has 3 states which are expressed in Eqs. (28)-(30). The parameters are available in [33]. The transient behavior of the model is shown in Figure 4.

$$\frac{dX}{dt} = -DX + \mu X, \qquad (28)$$

$$\frac{dS}{dt} = D(S_f - S) - \frac{1}{Y_{X/S}} \mu X, \qquad (29)$$

$$\frac{dP}{dt} = -DP + (\alpha\mu + \beta)X, \qquad (30)$$

$$\mu = \frac{\mu_m (1 - \frac{P}{P_m})}{K_m + S + \frac{S^2}{K_i}}.$$

The initial value of the states is $[5.9956 \ 5.0109 \ 19.1267]^T$. The nonlinear plant's data is achieved through sampling in the time interval 0 - 70 hr with a sampling time of $\Delta t = 0.25$ hr. The second order transfer function (G_{SO}) is used for linear approximation of the nonlinear system and NLM computation. System inputs are used in a span of -60% to $+60\%$ relative to equilibrium values. The NLM of the states of this system are indicated in Table 3.

Table 2. The results of Example 2 with different linear approximations

	G_{FO}		G_{SO}		G_{ZO}		Helbig et al.	Shastri et al.†
Output	$\theta^{\mathcal{U}_c}_{NOP}$	t_{cal} (sec)	$\theta^{\mathcal{U}_c}_{NOP}$	t_{cal} (sec)	$\theta^{\mathcal{U}_c}_{NOP}$	t_{cal} (sec)	$\theta^{\mathcal{U}_c}_{NOP}$	$\theta^{\mathcal{U}_c}_{NOP}$
c_A	0.2364	223.3753	0.2584	26.5446	0.2612	0.0754	0.2478	-
c_B	0.9883	431.8095	1.0000	43.3334	1.0000	0.0867	0.9912	1.0000
T	0.9854	244.6982	1.0000	43.4365	1.0000	0.0883	0.9921	1.0000
T_J	0.9857	244.2729	0.9995	41.3064	1.0000	0.0794	0.9957	1.0000

† Sign change in the gain of process around an operating point leads to the maximum nonlinearity.

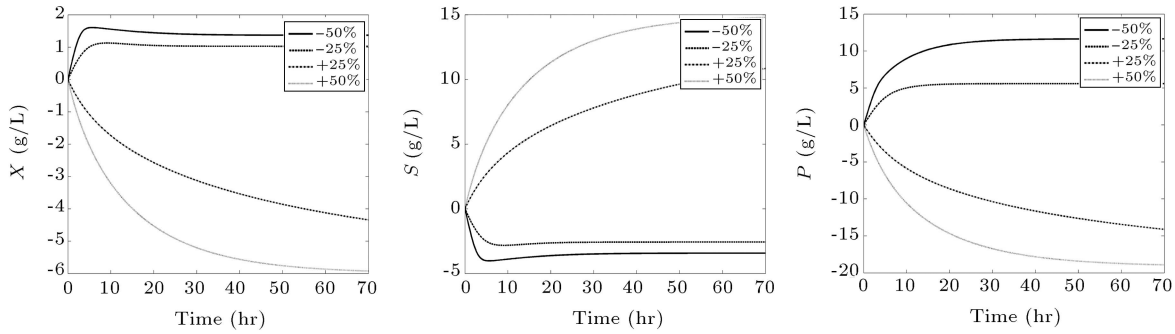

Figure 4. Transient behavior of nonlinear Fermentor's states.

Table 3. Nonlinearity measure of the continuous Fermentor's states.

Output	$\theta^{\mathcal{U}_c}_{NOP}$	t_{cal} (sec)
X	0.7020	22.7404
S	0.6993	24.5368
P	0.3706	38.6037

5.4. Example 4-1

The pH neutralization system is investigated. This system is a well known Wiener type nonlinear system which is investigated as a benchmark example of nonlinear systems in the literature. The description states for the transient behavior of the nonlinear system are expressed in Eqs. (31)-(33). The system parameters are adapted from [34]. The initial values of system states (x_0) are $[5.9956\ 5.0109\ 19.1267)]^T$, and the initial value of pH is 7. Figure 5 shows the measured output of the system, regarding three different inputs.

$$\frac{dW_{a4}}{dt} = \frac{1}{Ah}(q_1(W_{a1} - W_{a4}) + q_2(W_{a2} - W_{a4}) + q_3(W_{a3} - W_{a4})), \tag{31}$$

$$\frac{dW_{b4}}{dt} = \frac{1}{Ah}(q_1(W_{b1} - W_{b4}) + q_2(W_{b2} - W_{b4}) + q_3(W_{b3} - W_{b4})), \tag{32}$$

$$\frac{dh}{dt} = q_1 + q_2 + q_3 - C_v(h+z)^n. \tag{33}$$

The sampling procedure is performed in the time interval of 0-20 min, with sampling time of $\Delta t = 15$ sec. An approximation of a nonlinear system with the parallel second order transfer function (G_{SO}) is presented. The computation results for the pH neutralization system are indicated in Table 4. the NLM value is calculated

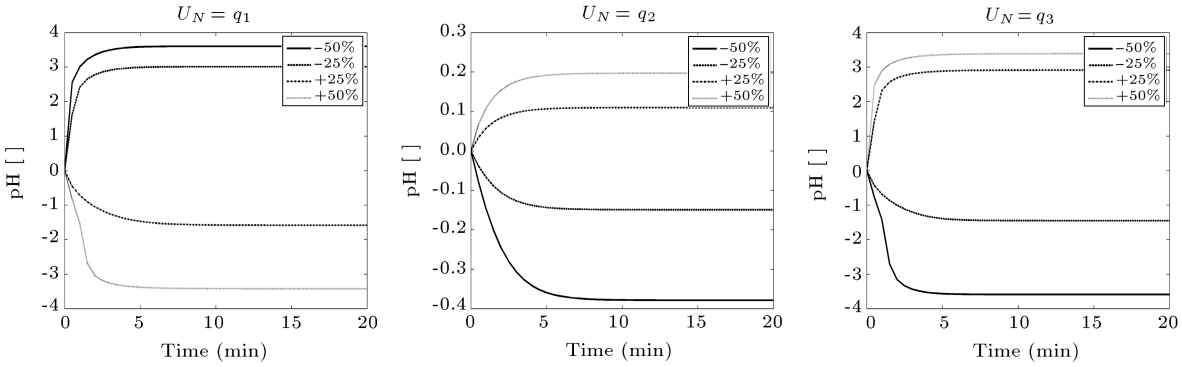

Figure 5. Transient behavior of the pH neutralization system in relative to 3 manipulating inputs.

Table 4. The nonlinearity measure of pH neutralization system's states relative to inputs.

Input	q_1		q_2		q_3	
Output	$\theta^{\mathcal{U}_c}_{NOP}$	t_{cal} (sec)	$\theta^{\mathcal{U}_c}_{NOP}$	t_{cal} (sec)	$\theta^{\mathcal{U}_c}_{NOP}$	t_{cal} (sec)
W_{a4}	0.3067	55.90	0.0332	38.07	0.2863	66.50
W_{b4}	0.3067	88.51	0.0332	48.01	0.2863	86.34
W_{h4}	0.0731	28.04	0.0194	30.16	0.0681	28.61
pH	0.5722	30.61	0.3858	29.05	0.5691	33.87

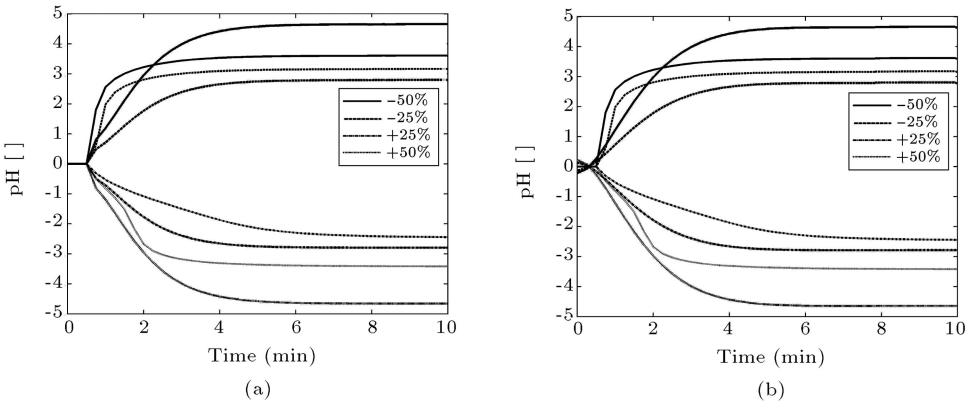

Figure 6. Response of the approximate second order linear system with (a) and without (b) considering time delay in optimization parameters, in comparison to the nonlinear outputs (gray curves correspond to nonlinear behavior).

and presented for the output (pH) and also the states of the system.

5.5. Example 4-2

In the pH neutralization process discussed previously, the time delay corresponding to the measurement was not considered. In this part, the same process, along with time delay in the measurement, is considered. The input-output pair of q_1-pH is studied. Sampling time is the same as in Example 4-1, and a dead time of 30 sec for pH measurement is considered. As illustrated in Figure 6(a), the optimized SOPDT can properly approximate the desired linear system, as well as the second order transfer function, without delay in Figure 6(b). In order to see the dead time properly in the linear model, the norm of the difference between two nonlinear and linear systems at the earlier times of simulation, is added to the objective function with a very small weight ($w = 0.005$). The results of the NLM value are approximately the same as Example 4-1, as recorded in Table 5. The estimated value of dead time by SOPDT is exactly the same as the actual process, i.e. 30 sec. In addition, NLM is calculated using G_{FO}.

According to the results illustrated in Table 5, the NLM values are almost equal in the case of Example 4-1 using G_{SO}, with and without dead time, and G_{FO}. Again, in the presence of dead time in the pH measurement, the NLM values show some variance, which is caused by the unmeasured dead time in the plant's input-output data. However, the SOPDT transfer function gives the same results when there is no dead time in the pH measurement.

Table 5. NLM of q_1-pH pair in pH neutralization system, with and without Dead Time (DT).

DT	0 sec	30 sec
G_{FO}	0.5753	0.6084
G_{SO} without DT	0.5722	0.5861
G_{SO} with DT	0.5722	0.5721

5.6. Example 5

In this example, the nonlinearity measurement of a high purity distillation column proposed by Skogestad & Morari [35] is studied. A distillation column is one of the most complicated systems in chemical engineering, regarding modeling and simulation, which is caused by the huge number of observable and non-observable state variables and system parameters. The reason why this system was simulated by a commercial dynamic simulator was to make its behavior similar to what happens in a real process without any simplifying assumption. The input-output data from simulation are used to compute the NLM of this system. A high purity distillation column is an important nonlinear process, which shows a huge change in the magnitude of the process gain, with respect to the sign change of the inputs. Modeling data for the distillation column are adapted from [35]. The input flow rate to the column is considered a manipulating variable, and the concentrations of the main product in the top and bottom of the column are considered as process outputs. The model is illustrated in Figure 7.

The results of stimulation are indicated in Figure 8 for distillate concentration (x_D). It is clear in Figure 8 that the gain of the system has a huge

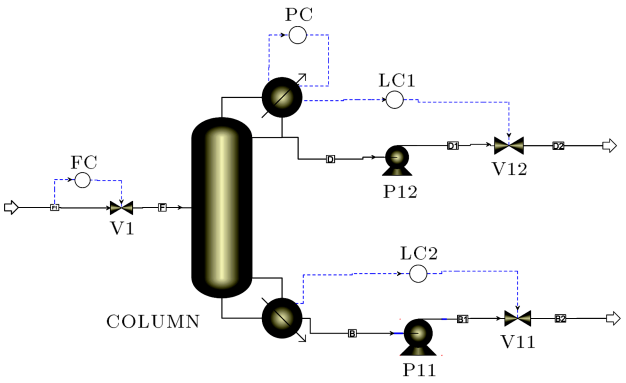

Figure 7. Two product distillation column of Skogestad and Morari [35], simulated by a commercial simulator.

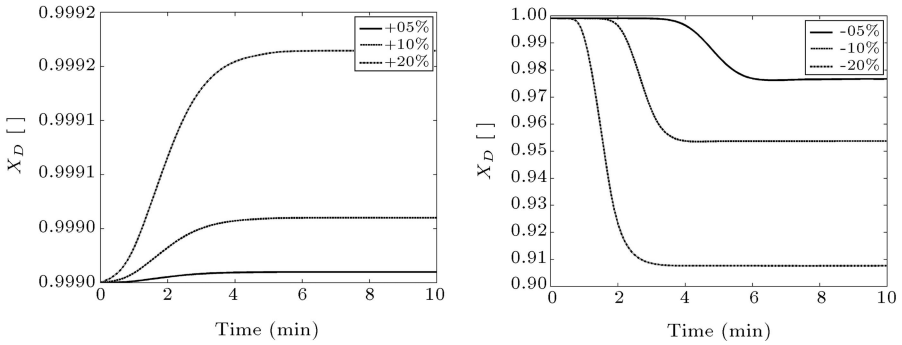

Figure 8. Transient behavior of distillation column for distillate product.

change against positive and negative stimulations. For instance, in the top of the column, x_D changes in the range of $0.999 - 1$ for positive stimulation, whereas there is no rigid limitation in the negative stimulation. Exactly the same behavior is observed in the concentration at the bottom of the column (x_B). At the bottom of the column, when a positive change is applied to the input flow rate, x_B can change in a wide range, whilst in a negative stimulation, x_B can vary in a limited range of $0 - 0.001$. This behavior leads to the huge nonlinearity of the high purity distillation column, as the NLM calculation of $\theta^{\mathcal{U}_c}_{N_{OP}} = 1$ confirms. Furthermore, as seen in Figure 8, there is a time varying delay in the input-output behavior of the system which is induced by the huge number of state variables of the system with the changing parameters. However, the main cause of the severe nonlinear behavior of this system is the intense changes of the process gain, since the use of the SOPDT transfer function did not change the result of NLM calculation.

5.7. Example 6

This example is used to show the effect of the relative order of the system under study, on the NLM calculation. A system of two tanks (like the proposed CSTR in Example 1), which are connected in series and are cooled by a cooling stream, is discussed. This system is higher order in comparison to the system in Example 1, and the first tank is exactly the CSTR illustrated in Example 1. The model equations and parameters are used from [36]:

$$\frac{dc_{A1}}{dt} = \frac{q}{V_1}(c_{Af} - c_{A1}) - k_0 c_{A1} \exp(-\frac{E}{RT_1}), \quad (34)$$

$$\frac{dT_1}{dt} = \frac{q}{V_1}(T_f - T_1) + \frac{(-\Delta H)k_0 c_{A1}}{\rho C_p} \exp(-\frac{E}{RT_1}) + \frac{\rho_c C_{pc}}{\rho C_p V_1} q_c (1 - \exp(-\frac{hA_1}{q_c \rho_c C_{pc}}))(T_{cf} - T_1), \quad (35)$$

$$\frac{dc_{A2}}{dt} = \frac{q}{V_2}(c_{A1} - c_{A2}) - k_0 c_{A2} \exp(-\frac{E}{RT_2}), \quad (36)$$

$$\frac{dT_2}{dt} = \frac{q}{V_2}(T_1 - T_2) + \frac{(-\Delta H)k_0 c_{A2}}{\rho C_p} \exp(-\frac{E}{RT_2}) + \frac{\rho_c C_{pc}}{\rho C_p V_2} q_c (1 - \exp(-\frac{hA_2}{q_c \rho_c C_{pc}})) .(T_1 - T_2 + \exp(-\frac{hA_1}{q_c \rho_c C_{pc}})(T_{cf} - T_1)), \quad (37)$$

where the initial conditions are $c_{A1,0} = 8.823 \times 10^{-2}$ $\frac{\text{mol}}{\text{L}}$, $T_{1,0} = 441.22$ K, $c_{A2,0} = 5.293 \times 10^{-3}$ $\frac{\text{mol}}{\text{L}}$, and $T_{2,0} = 449.47$ K. The manipulating input is the coolant water flow rate ($u_{N,in} = q_c$), and $u_{N,s} = 100$ $\frac{\text{L}}{\text{min}}$. The input region is considered to be similar to that used in Example 1. The time interval of simulation is 0-20 min, with the sampling time of $\Delta t = 0.1$ min. The output of this system is the concentration of component A in the second tank (c_{A2}). The transient behavior of this system is shown in Figure 9.

As predicted from comparison of the input-output behavior of the systems with one and two CSTRs, respectively, in Figures 1 and 9, the NLM values are close. The nonlinear system introduced in this example is a little more nonlinear than the system introduced

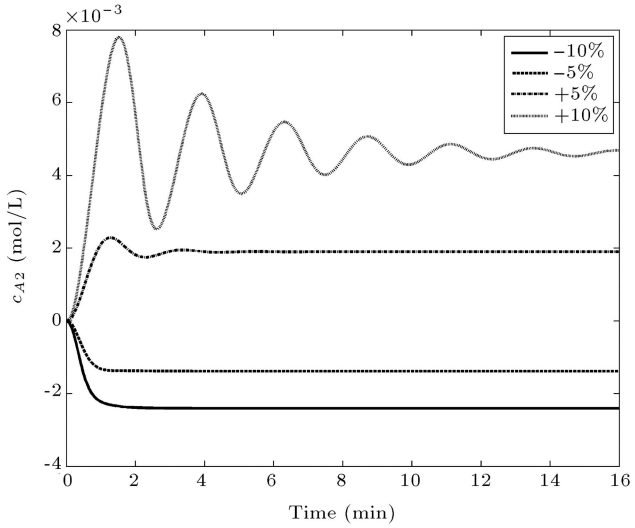

Figure 9. Transient behavior of c_{A2} in the two CSTR system of Example 6.

in Example 1 in which one CSTR existed with the relative order of one. The NLM value of this system is $\theta^{\mathcal{U}_c}_{N_{OP}} = 0.3522$.

5.8. Example 7

The nonlinearity assessment of Sorensen's physiological model of T1DM is implemented. The number of state variables in this nonlinear system is numerous. Sorensen's model consists of 19 states in three sub-models of Glucose, Insulin and Glucagon, with 11, 7, and 1 states, respectively. The manipulating input of this model for a T1DM patient is the injection rate of insulin, and the measured output is the plasma glucose concentration. The describing equations and the physiological parameters of the model are given in [4]. It is well known that the large number of nonlinear or even purely linear state variables would cause a time delay in the outputs of a system. This time delay in the case of purely linear states would be the cause of the nonlinear behavior of the system, whereas the main system is linear. The values of the input and output at the equilibrium point are $u_s = 25\frac{\text{mU}}{\text{min}}$ and $G_{H,s} = 85\frac{\text{mg}}{\text{dL}}$, respectively. The manipulating input is stimulated in the range of $|u_N| = 20\ \frac{\text{mU}}{\text{min}}$ in the vicinity of an equilibrium point. The simulations are implemented in the time interval of 0-500 min, with sampling time of $\Delta t = 1$ min. The value of NLM is computed as $\theta^{\mathcal{U}_c}_{N_{OP}} = 0.1682$. The system is not so much nonlinear in the sense of this criterion, as is clear in Figure 10. The nonlinearity of the nonlinear states of this system is caused by $tanh(.)$ terms which describe metabolic rates. Since the order of magnitude of these rate terms is small compared with the other terms in the states of this equation, the nonlinearity of the total system is not affected by these nonlinearities.

Again, the value of NLM decreases when SOPDT is used instead of G_{SO}, as the approximating linear system. The NLM is calculated as $\theta^{\mathcal{U}_c}_{N_{OP}} = 0.1589$ in this manner, regarding an estimated dead time of $DT = 11$ min. As stated before, this large system with nonlinear state variables, shows a totally linear behavior.

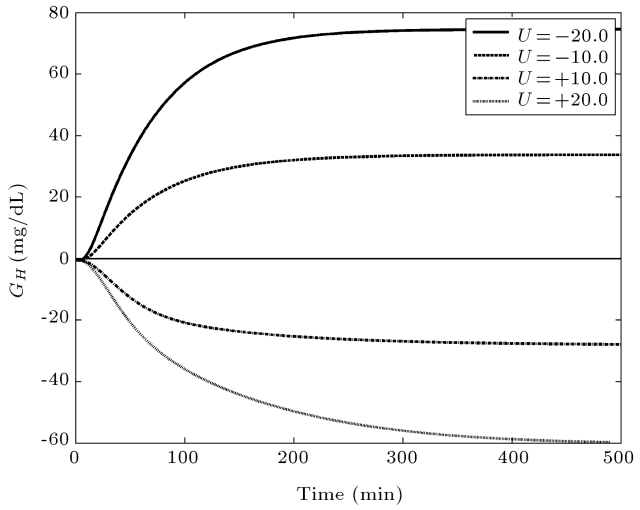

Figure 10. Response of deviation plasma glucose level (in $\frac{\text{mg}}{\text{dL}}$) with respect to 4 step stimulations of the insulin injection rate (in $\frac{\text{mU}}{\text{min}}$).

6. Conclusion

In this paper, the results of simulation and NLM computation in Examples 1 and 2 show that the suggested computed values using two approximations in this study are in reasonable agreement with the results reported by other researchers. The number of parameters in the suggested linear approximation is less than the others in the literature. Therefore, the required amount of computation for solving the optimization problems is less and consequently, the required calculation time will decrease. Computation of the NLM of a system ($\theta^{\mathcal{U}_c}_{N_{OP}}$) using a second order linear transfer function (G_{SO}) is about 7.4 times faster than the approach in which the weighted summation of first order linear transfer functions (G_{FO}) is used. Moreover, estimation of the NLM of a system using zero order comparison functions is 466.0 and 3447.68 times faster compared to using G_{SO} and G_{FO}. Additionally, there is no restriction on generating oscillations in the use of G_{SO} rather than G_{FO} in which the number of oscillations is limited to the number of first order transfer functions (the number of transfer functions is proportional to the number of optimization parameters, which geometrically increases computational cost). The dead time estimation is also observed in the use of SOPDT.

In this research, seven standard and frequently used benchmarks of nonlinear chemical processes have been investigated. The case studies contain various types of nonlinear system: mildly to highly nonlinear systems, systems with relative degrees of one, two and more, systems with small to large numbers of states, systems with a drastic change in the process gain value, and systems with a sign change of the process gain. The results obtained show that the main issue affecting the nonlinearity measure is the input-output behavior of the system, since the nonlinearity measure was originally defined based on the input-output trend of the system. Hence, various issues, like the relative order and the number of states, cannot be used as a major decision criterion in the nonlinearity assessment of the process. According to the results obtained, the nonlinearity of systems increases in the following order: Sorensen's T1DM model in Example 7, the CSTR reactor in Example 1, two serially connected CSTRs in Example 6, pH neutralization systems in Example 4, the fermentation system in Example 3, the Van de

Vusse system in Example 2, and high purity distillation columns in Example 5.

Due to the fast estimation of the NLM via the proposed method, it can be employed in an online manner. For instance, it can be used in the online selection or estimation of linear models in the identification and control of nonlinear systems.

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