Local parameter identifiability of large-scale nonlinear models based on the output sensitivity covariance matrix ⋆

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Abstract: The use of first-principle models is motivated by the potential of detailed information available as well as their versatility. Therefore, it is important to keep these models up to date so the models represent accurate enough the processes at hand. However, most of these models are nonlinear with a large number of states and parameters but with a relatively low number of measured outputs. This lack of measurements hinders the possibility to estimate all of the parameters present in the model. In this work, parameter identifiability of large-scale nonlinear models is explored using the empirical output controllability covariance matrix approach. This empirical covariance matrix is used to extract the output sensitivity matrix of the model to assess parameter identifiability. The advantages of the proposed methods are discussed while different sensitivity indexes are evaluated to draw sound conclusions on the parameter ranking results. A large-scale reactive batch distillation process simulation is used as a demonstrator.

Keywords: Empirical covariance matrix, Empirical gramian, Sensitivity analysis, Identifiability, Output controllability

1. INTRODUCTION

The use of mathematical models to make decisions, monitor, optimize, and/or control a process, encompasses the so-called digital twin paradigm (Grieves and Vickers (2017)) or master model paradigm (Backx et al. (2000)). This particular setting requires constant communication between the plant and the model to perform periodic alignment of the model outputs and plant measurements if needed. Despite the clear advantages these models offer, they tend to be very complex with a large number of variables involved; hence, keeping these models up to date is a challenging task. Under the assumption of no process undermodeling, the discrepancies between the model and the plant can be minimized/or eliminated performing parameter estimation. Nevertheless, the processes represented by a digital twin usually have a limited number of measured outputs. This constraints the amount of information that can be extracted from the process. Moreover, parameters might not be all identifiable from the available inputs and outputs. Thus, it must be first determined which parameters can be indeed estimated with relatively high certainty. In this sense, parameter output sensitivity is an attractive approach because not only does it allow to assess parameter identifiability, but also the design of insensitive control (Okura and Fujimoto (2016)), input design for parameter identification (Stigter and Peeters (2007)), model parameter reduction (Sun and Hahn (2006)), etc. Additionally, the underlying mathematical model can be very complex, making analytical approaches for the analysis of parameter sensitivity and identifiability intractable. To deal with such complexity, numerical approaches can be deployed to analyze local parametric properties of the model. A comprehensible review on experiment design and parameter estimation can be found in Franceschini and Macchietto (2008). In this work, the empirical output parameter sensitivity covariance matrix is extracted from the empirical output controllability covariance matrix to assess parameter identifiability of nonlinear models. A comparison to the identifiability covariance matrix based on the observability covariance matrix is done, where similarities and differences are discussed. Furthermore, a discussion on different sensitivity indexes is presented to show that the parameter ranking changes drastically depending on the sensitivity index used. The approach is based upon the works of Hahn et al. (2003) and Sun and Hahn (2006), and is extended for stable systems and systems that do not possess fixed-point steady states.

The paper is organized as follows: Section 2 introduces empirical gramians and empirical covariance matrices. Section 3 presents the empirical output sensitivity covariance matrix and the identiafibility covariance matrix, as well as their connection to parameter identifiability. Section 4 explores different sensitivity indexes that account for the sensitivity magnitude of each output with respect to each parameter. Subsequently, Section 5 treats the case study of a large-scale reactive batch distillation column in which a set of parameters is assessed for identifiability using the empirical output sensitivity covariance matrix. Finally, Section 6 contains the conclusions of this work.

2. EMPIRICAL GRAMIANS AND SENSITIVITY COVARIANCE MATRICES

Let $\mathcal{S}$ be a dynamical system, and $\mathcal{M}(\cdot)$ be a model structure. Furthermore, $\mathbf{p} \in \mathbb{P} \subseteq \mathbb{R}^{n_p}$ be a vector of parameters, where $\mathbb{P}$ is the feasible parameter space, and $\mathcal{M}(\mathbf{p})$ be the specific model obtained when the parameters take the values $\mathbf{p}$. In this work, we restrict our attention to general nonlinear ordinary differential equation (ODE) models of the form

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$$\mathcal{M}(\mathbf{p}) = \begin{cases} \dot{x}(t,\mathbf{p}) = f(x(t,\mathbf{p}),u(t),\mathbf{p}), \;\; x(0) = x_0 \\ y(t,\mathbf{p}) = h(x(t,\mathbf{p})) \\ \dot{\mathbf{p}} = \mathbf{0} \end{cases} \tag{1}$$

where $t \in \mathbb{R}_{[0,t_f]}$, $x(t,\mathbf{p}) \in \mathbb{R}^{n_x}$ is the vector of states, $u(t) : [0,t_f] \mapsto \mathbb{R}^{n_u}$ is the known input vector, $y(t,\mathbf{p}) \in \mathbb{R}^{n_y}$ is the output vector.

Two important properties of the model structure $\mathcal{M}(\mathbf{p})$ described in (1) is controllability (reachability) and observability properties. Each of these properties can be evaluated using the controllability and observability gramian matrices. In the linear case, these matrices are given by

$$W_{C,\text{lin}} = \int_0^\infty e^{At} B B^\top e^{A^\top t} \; \mathrm{d}t \tag{2}$$

$$W_{O,\text{lin}} = \int_0^\infty e^{A^\top t} C^\top C e^{At} \; \mathrm{d}t \tag{3}$$

Another important gramian is the output controllability gramian. The output controllability gramian contains information about the input-output map of a particular model. Furthermore, it is advantageous for large scale systems where the number of states is too large to compute the output controllability gramian, as it only requires the measured outputs. Additionally, it also allows to analyze the effect of initial conditions and inputs on the output. Therefore, the output controllability encodes both the controllability and observability properties of a model. The output controllability for a linear model as described in Casadei et al. (2018) is given by

$$W_{C^o,\text{lin}} = \int_0^\infty C e^{At} B B^\top e^{A^\top t} C^\top \; \mathrm{d}t \tag{4}$$

However, the gramians in (2), (3), and (4) can only be computed for linear models. In the case of nonlinear model, empirical gramians can be calculated, but these are restricted to Lyapunov-stable control-affine nonlinear models (Lall et al. (2002)). Therefore, an extension to general nonlinear models was made by Hahn et al. (2003) to generate expressions similar to the controllability and observability gramians, namely the controllability covariance and observability covariance matrices. These covariance matrices contain the controllability and observability gramians as special cases.

Definition 1. (Empirical controllability covariance matrix). Let the system given by (1) be stable, and introduce the following sets

$$\begin{aligned} T^{n_u} &= \{T_1, \cdots, T_r \in \mathbb{R}^{n_u \times n_u}, T_i^\top T_i = \mathbf{I}_{n_u}, i = 1, \cdots, r\} \\ M &= \{c_1, \cdots, c_s \in \mathbb{R}, c_i > 0, i = 1, \cdots, s\} \\ E^{n_u} &= \{e_1, \cdots, e_{n_u};\ \text{standard unit vectors in } \mathbb{R}^{n_u}\} \end{aligned}$$

where r is the number of matrices for perturbation directions, s is the number of different perturbation sizes for each direction, and n_u is the number of inputs.

The state-controllability covariance matrix is defined as

$$W_C = \sum_{\ell=1}^{n_u} \sum_{m=1}^{r} \sum_{n=1}^{s} \frac{1}{src_n^2} \int_0^\infty \Phi^{\ell mn}(t) \; \mathrm{d}t \tag{5}$$

where $\Phi^{\ell mn}(t) \in \mathbb{R}^{n_x \times n_x}$ is computed as $\Phi^{\ell mn}(t) = (x^{\ell mn}(t) - x_{\text{ss}}^{\ell mn})(x^{\ell mn}(t) - x_{\text{ss}}^{\ell mn})^\top$, and $x^{\ell mn}$ is the state corresponding to the input $u(t) = c_n T_m e_i v(t) + u_{\text{ss}}$, with $v(t)$ denoting the nature of the input signal, and x_{ss} and u_{ss} are the state and input steady-state values.

Definition 2. (Empirical observability covariance matrix). Let the system given by (1) be stable, and introduce the following sets

$$\begin{aligned} T^{n_x} &= \{T_1, \cdots, T_r \in \mathbb{R}^{n_x \times n_x}, T_i^\top T_i = \mathbf{I}_{n_x}, i = 1, \cdots, r\} \\ M &= \{c_1, \cdots, c_s \in \mathbb{R}, c_i > 0, i = 1, \cdots, s\} \\ E^{n_x} &= \{e_1, \cdots, e_{n_x};\ \text{standard unit vectors in } \mathbb{R}^{n_x}\} \end{aligned}$$

where r is the number of matrices for perturbation directions, s is the number of different perturbation sizes for each direction, and n_x is the number of states.

The observability covariance matrix is defined as

$$W_O = \sum_{m=1}^{r} \sum_{n=1}^{s} \frac{1}{src_n^2} \int_0^\infty T_m \Psi^{mn}(t) T_m^\top \; \mathrm{d}t \tag{6}$$

where $\Psi^{mn}(t) \in \mathbb{R}^{n_x \times n_x}$ is computed as $\Psi_{ij}^{mn}(t) = (y^{imn}(t) - y_{\text{ss}}^{imn})(y^{jmn}(t) - y_{\text{ss}}^{jmn})^\top$, and y^{imn} is the output corresponding to the initial state $x(0) = c_n T_m e_i + x_{\text{ss}}$, and y_{ss}^{imn} is steady-state output that the system will reach after such perturbation.

In Definitions 1 and 2, the state-controllability and observability covariance matrices are defined for stable systems, ensuring that states, inputs, and outputs reach the steady-state. However, these particular definitions fail in the case of processes that do not posses steady-states, e.g. unstable systems or batch processes. Moreover, this type of systems might not be able to run for an indefinite amount of time, which is the case of batch processes. Therefore, we use the generalization introduced by Saltik et al. (2016) to replace x_{ss} and u_{ss} by nominal trajectories and compute a finite-time covariance matrices with respect to nominal trajectories. On the other hand, the number of states can be too large in a large-scale system for the computation of the empirical state-controllability covariance matrix to be feasible. Thus, we can construct the empirical output-controllability covariance matrix (EOCCM) using the output signals instead of the states, since the number of measured outputs of a system is generally lower than that of the states. Thus, the proposed finite-time EOCCM is

$$\bar{W}_{C^o} = \sum_{\ell=1}^{n_u} \sum_{m=1}^{r} \sum_{n=1}^{s} \frac{1}{src_n^2} \int_0^{t_f} \bar{\Phi}^{\ell mn}(t) \; \mathrm{d}t \tag{7}$$

where $\bar{\Phi}^{\ell mn}(t) \in \mathbb{R}^{n_y \times n_y}$ is computed as $\bar{\Phi}^{\ell mn}(t) = (y^{\ell mn}(t) - \bar{y})(y^{\ell mn}(t) - \bar{y})^\top$, and $y^{\ell mn}$ is the state corresponding to the input $u(t) = c_n T_m e_i v(t) + \bar{u}$, with $\bar{y}$ and $\bar{u}$ the state and input nominal trajectory respectively.

The EOCCM contains information of the input-output information directly, which also contains information on the observability and controllability of the system. It can be used as a general tool for system analysis rather than the separate empirical controllability and observability matrices .

3. COMPUTATION OF PARAMETRIC SENSITIVITIES USING COVARIANCE MATRICES

Another important property of the model structure $\mathcal{M}(\mathbf{p})$ is parameter identifiability. In its most general form, identifiability accounts to determine if the map $\Phi : \mathbf{p} \mapsto \mathcal{M}(\mathbf{p})$

is injective, i.e. the map has one-to-one correspondence. However, in most practical settings, the only measured variable that provides information about the model behavior is the output y. This implies that the entire model structure $\mathcal{M}(\mathbf{p})$ can be replaced by the input/output map $\Gamma(\mathbf{p}) : \mathcal{U} \mapsto \mathcal{Y}$, whose domain is the input space $\mathcal{U} \subset \mathbb{R}^{n_u}$ containing admissible input signals u, and codomain is the output space $\mathcal{Y} \subset \mathbb{R}^{n_y}$ with output signals y as elements. The structural identifibiality analysis accounts to verifying injectivity of the map $\bar{\Phi} : \mathbf{p} \mapsto \Gamma(\mathbf{p})$. We shall now formalize this by the following definitions for ODE models by Ljung and Glad (1994):

Definition 3. (Global identifiability). A model $\mathcal{M}(\mathbf{p})$ given by (1) is globally identifiable if for any admissible input $u \in \mathcal{U}$, and initial condition x_0

$$\begin{aligned} \exists x_0, \exists u, \forall \mathbf{p}, \mathbf{p}^* \in \mathbb{P} : \quad & y(x(t), u(t), \mathbf{p}) = y(x(t), u(t), \mathbf{p}^*) \\ & \Rightarrow \mathbf{p}_i = \mathbf{p}_i^* \ \ \forall \mathbf{p}^* \in \mathbb{P} \end{aligned} \tag{8}$$

holds.

Definition 4. (Local identifiability). Let $N(\mathbf{p}_0)$ define the neighborhood of a particular point $\mathbf{p}_0$ in the parameter space $\mathbb{P}$. A model $\mathcal{M}(\mathbf{p})$ given by (1) is locally identifiable at $\mathbf{p}^*$ if for any admissible input $u \in \mathcal{U}$, and initial condition x_0

$$\begin{aligned} \exists x_0, \exists u : \quad & y(x(t), u(t), \mathbf{p}) = y(x(t), u(t), \mathbf{p}^*) \\ & \Rightarrow \mathbf{p}_i = \mathbf{p}_i^* \ \ \forall \mathbf{p} \in N(\mathbf{p}^*) \subset \mathbb{P} \end{aligned} \tag{9}$$

holds.

Identifiability can be tested in many ways, but most methods can be classified in two categories: structure-based and simulation-based. The former deals with the identifiability problem using a formal mathematical approach, whose implementation requires symbolic packages, while the latter takes on a numerical approach. The structural approach allows for a more in depth assessment, such as global and/or local identifiability as well as calculation of identifiable parameter combinations. However, it is only suitable for small-scale systems with few parameters and states due to the curse of dimensionality. Conversely, simulation-based approaches can deal with large-scale systems at the expense of obtaining just local results, i.e. in the vicinity of the operating conditions. In this context, we can use the covariance matrix to test for parameter identifiability. Two approaches can be devised for parameter identifiability Himpe and Ohlberger (2013): the empirical sensitivity covariance matrix and the empirical identifiability covariance matrix. The two matrices are calculated as follows:

Extended output controllability covariance matrix

Around a nominal trajectory (or point in the case of systems with steady-states), the system (1) can be represented by a linear system that approximates the behavior in the neighborhood of the aforementioned trajectory. Hence,

$$\begin{aligned} \delta\dot{x} &= J_{f,x}\delta x + \sum_{i=1}^{n_p} J_{f,\mathbf{p}_i}\delta\mathbf{p}_i + J_{f,u}\delta u \\ \delta y &= J_{h,x}\delta x \end{aligned} \tag{10}$$

where $\delta x = x - \bar{x}$, $\delta\mathbf{p}_i = \mathbf{p}_i - \bar{\mathbf{p}}_i$, and $\delta y = y - \bar{y}$, and $\delta u = u - \bar{u}$, with $\bar{(\cdot)}$ represents the nominal value. $J_{f,x}$, $J_{f,u}$, $J_{h,x}$, and $J_{h,u}$ are the Jacobian matrices of functions f and h with respect to the states and inputs, respectively. $J_{f,\mathbf{p}}$ is the Jacobian matrices of the function f with respect to the i-th parameter.

Notice, that at each point of the linearization, the system can be realized as a sum of the effects of the inputs and parameters on each through the states to the outputs (Sun and Hahn (2006)). Therefore, the output controllability covariance matrix is given by

$$\bar{W}_{C^o} = \bar{W}_{C^o}[u] + \sum_{i=1}^{n_p} \bar{W}_{C^o}[\mathbf{p}_i] \in \mathbb{R}^{n_y \times n_y} \tag{11}$$

The right most terms of (11) correspond to each output sensitivity covariance matrix whose representation is given by

$$\bar{W}_{C^o}[\mathbf{p}_i] = \begin{bmatrix} \left(\frac{\partial\delta y_1}{\partial\delta\mathbf{p}_i}\right)\left(\frac{\partial\delta y_1}{\partial\delta\mathbf{p}_i}\right)^\top & \cdots & \left(\frac{\partial\delta y_1}{\partial\delta\mathbf{p}_i}\right)\left(\frac{\partial\delta y_{n_y}}{\partial\delta\mathbf{p}_i}\right)^\top \\ \vdots & \ddots & \vdots \\ \left(\frac{\partial\delta y_{n_y}}{\partial\delta\mathbf{p}_i}\right)\left(\frac{\partial\delta y_1}{\partial\delta\mathbf{p}_i}\right)^\top & \cdots & \left(\frac{\partial\delta y_{n_y}}{\partial\delta\mathbf{p}_i}\right)\left(\frac{\partial\delta y_{n_y}}{\partial\delta\mathbf{p}_i}\right)^\top \end{bmatrix}$$

The information contained in each of the matrices of the sum shown in (11) corresponds to the sensitivity of the output with respect to each individual parameter of the model, i.e. the total variation of the output to changes in each parameter of the model. Notice that there are as many sensitivity matrices as parameters. Moreover, each of the sensitivity matrices provides information about the interaction between all outputs with respect to the one parameter, which is helpful in determining the outputs that should be measured. The construction of the sensitivity covariance matrix presented in this work differs from proposed one in Sun and Hahn (2006), where the development is based on the observability covariance matrix.

Extended observability covariance matrix

Let the system (1) be represented by a linear system around a nominal trajectory. Thus,

$$\begin{aligned} \begin{bmatrix} \delta\dot{x} \\ \delta\dot{\mathbf{p}} \end{bmatrix} &= \begin{bmatrix} J_{f,x} & J_{f,\mathbf{p}} \\ \mathbf{0} & \mathbf{0} \end{bmatrix} \begin{bmatrix} \delta x \\ \delta\mathbf{p} \end{bmatrix} + \begin{bmatrix} J_{f,u} \\ \mathbf{0} \end{bmatrix} \delta u, \quad \delta x(0) = \delta x_0 \\ \delta y &= [J_{h,x} \ \ \mathbf{0}] \begin{bmatrix} \delta x \\ \delta\mathbf{p} \end{bmatrix} \end{aligned} \tag{12}$$

where δx, $\delta\mathbf{p}$, δy, δu, $J_{f,x}$, $J_{f,\mathbf{p}}$, J_{f_u}, and $J_{h,x}$ are defined as before. The observability matrix has the form (Singh and Hahn (2005), Saltik et al. (2016))

$$\bar{W}_O^i = \begin{bmatrix} \bar{W}_O^i[x_0] & \bar{W}_O^i[x_0, \mathbf{p}] \\ \bar{W}_{O,}^{i^\top}[x_0, \mathbf{p}] & \bar{W}_O^i[\mathbf{p}] \end{bmatrix} \tag{13}$$

The lower diagonal block corresponds to the matrix of output sensitivity covariance matrix, whose representation is given by

$$\bar{W}_O^i[\mathbf{p}] = \begin{bmatrix} \left(\frac{\partial \delta y_i}{\partial \delta \mathbf{p}_1}\right)^\top \left(\frac{\partial \delta y_i}{\partial \delta \mathbf{p}_1}\right) & \cdots & \left(\frac{\partial \delta y_i}{\partial \delta \mathbf{p}_1}\right)^\top \left(\frac{\partial \delta y_i}{\partial \delta \mathbf{p}_{n_p}}\right) \\ \vdots & \ddots & \vdots \\ \left(\frac{\partial \delta y_i}{\partial \delta \mathbf{p}_{n_p}}\right)^\top \left(\frac{\partial \delta y_i}{\partial \delta \mathbf{p}_1}\right) & \cdots & \left(\frac{\partial \delta y_i}{\partial \delta \mathbf{p}_{n_p}}\right)^\top \left(\frac{\partial \delta y_i}{\partial \delta \mathbf{p}_{n_p}}\right) \end{bmatrix}$$

where $i \in \{1, 2, \cdots, n_y\}$. Notice that in this case, the sensitivity matrix is part of the extended observability covariance matrix, Himpe and Ohlberger (2013) defined the identifiability covariance matrix $\bar{W}_I^i$ as the lower-diagonal block Schur complement. Therefore

$$\bar{W}_I^i[\mathbf{p}] = \bar{W}_O^i[\mathbf{p}] - \bar{W}_O^{i^\top}[x_0, \mathbf{p}] \left(\bar{W}_O^i[x_0]\right)^{-1} \bar{W}_O^i[x_0, \mathbf{p}] \quad (14)$$

However, it is usually sufficient to approximate the empirical identifiability covariance matrix by the lower right block of (13) (Himpe (2018))

$$\bar{W}_I^i[\mathbf{p}] \approx \bar{W}_O^i[\mathbf{p}] \quad (15)$$

Note that that there are as many covariance matrices as outputs, and each of these matrices provides information on the interactions of all parameters with respect to one output. Although both sensitivity representations $\bar{W}_{C^o}[\mathbf{p}_i]$ and $\bar{W}_O^i[\mathbf{p}]$ might seem similar, they provide slightly different information. However, it is possible to obtain the same information from both matrices with the appropriate manipulations. Furthermore, the construction of the sensitivity covariance matrix from the output controllability perspective can be exploited to explore the influence of the input in the sensitivity analysis, which is relevant in input design for parameter estimation. Moreover, these covariance matrices are analogous to the Hessian matrix used in the Gauss-Newton method for parameter estimation.

4. INDEXES TO ASSESS LOCAL IDENTIFIABILITY

Another important factor when computing a ranking of parameters is the choice of a sensitivity index, which allows to quantify the influence of a parameter over the set of outputs. Several indexes have been used in literature to perform this assessment as discussed in Singh and Hahn (2005). We present the most common indexes used in literature.

Trace: This index measures the total contribution of a particular parameter over all of the outputs. From a parameter estimation point of view, the trace of the EOSCM gives a measure of the total uncertainty magnitude or total variation of a given parameter.

$$\iota_1^{S_i} = \text{trace}\left(\bar{W}_{C^o,}[\mathbf{p}_i]\right) \quad (16)$$

A modification can be performed to this measure to obtain the average variations of the outputs with respect to the parameters.

$$\iota_2^{S_i} = \frac{1}{n_y}\text{trace}\left(\bar{W}_{C^o,}[\mathbf{p}_i]\right) \quad (17)$$

Determinant: This index measures the linear independence of parameter sensitivity between output channels. If the determinant is small, then the parameters generate similar (linear dependent) variations in the outputs that are indistinguishable from one another. Mathematically, the determinant of the EOSCM provides a measure of the volume of the uncertainty or the generalized variation of a given parameter.

$$\iota_3^{S_i} = \det\left(\bar{W}_{C^o,}[\mathbf{p}_i]\right) \quad (18)$$

Analogously, a modification proposed by Müller and Weber (1972), provides an average magnitude of the volume of the uncertainty.

$$\iota_4^{S_i} = \sqrt[n_y]{\det\left(\bar{W}_{C^o,}[\mathbf{p}_i]\right)} \quad (19)$$

The particular use of these indexes in the context of parametric sensitivity is to determine whether a parameter is identifiable or not. The notion of a large or small trace (resp. determinant) is ambiguous. Although there is no straightforward way of choosing it, we propose to use a ratio between the total variation (trace) and the generalized variation (determinant) to assess identifiability. This ratio allows us to confine these values to the range $[0, 1]$. By the Arithmetic mean - Geometric mean (AM-GM) inequality, we have that

$$\iota_5^{S_i} = \frac{\iota_4^{S_i}}{\iota_2^{S_i}} \in [0, 1] \quad (20)$$

This value represents the ratio between the generalized variation to the total variation. A number close to 1 corresponds with high parametric sensitivity with low correlation between outputs, whereas a number close to 0 implies that the output is not sensitive to parameter variations or that induces high correlation between outputs.

Condition number: This index provides information about the posedness of the matrix. A large condition number implies that the matrix is ill-conditioned, thus, some directions of sensitivities are small or linearly dependent. On the other hand, a small condition number means that the matrix is well-conditioned, and that all sensitivity directions have similar magnitudes and are independent. This index plays an important role in numerical approaches for gradient-based parameter estimation, to select parameters that make the problem well-posed.

$$\iota_6^{S_i} = \frac{\lambda_{\max}(\bar{W}^e_{C^o,}[\mathbf{p}_i])}{\lambda_{\min}(\bar{W}^e_{C^o,}[\mathbf{p}_i])} \in [1, \infty) \quad (21)$$

In this work, we will use

$$\iota_7^{S_i} = \frac{1}{\iota_6^{S_i}} \in (0, 1] \quad (22)$$

The values close to 1 will represent identifiable parameters, and values close to 0 will represent condition number values that are very large; thus, unidentifiable parameters.

In general, there is no 'magical' index that will provide all the necessary information. Each of the indexes discussed above provide different pieces of identifiability information. The trace is a good measure if the sole interest is in the total contribution of the parameters on each output. However, the trace does not provide information about the correlation between sensitivities, which is well-captured by the determinant. Nevertheless, it does not allow to conclude on the total variation of the sensitivities. Analogously, the condition number provides information on the conditioning of the matrix without providing explicit values for variation or correlation in the data. The best option for sensitivity index will depend on the particular application or the information sought.

5. EXAMPLE: REACTIVE BATCH DISTILLATION

The case study used in this work is an industrial reactive batch distillation (RBD) process. The process consists of a reactor at the bottom acting also as a reboiler, and a standard tray distillation column on top. In the reactor, a series of chemical equilibrium reactions take place to produce the polymer and water as a by-product. The latter is then removed through the distillation column, constantly shifting the equilibrium to generate more desired final product. The measured outputs are the temperature and pressure in the reactor stage, and the input variables are the incoming jacket oil temperature, the pressure valve travel and the reflux flow. The process consists of 8 stages, and the reaction is only limited to the reactor stage where 10 chemical species interact in 16 chemical reactions. This amounts to a total of 90 differential equations, and over 470 algebraic equations. A schematic of the process in illustrated in Figure 1.

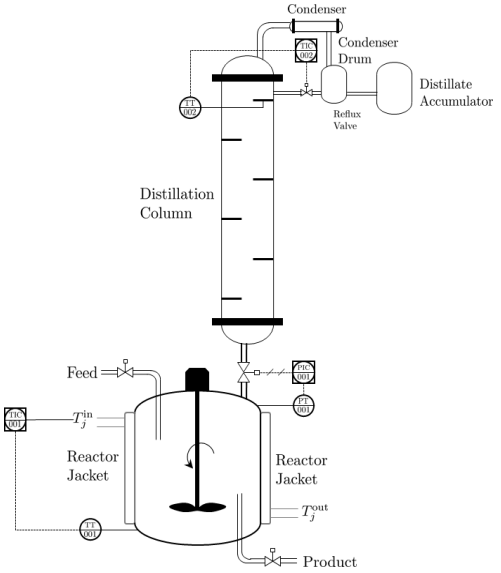

Fig. 1. Schematic representation of the RBD.

Molar Balance: Reactor stage

$\forall i = 1, \cdots, S, \;\; j = 8$

$$\frac{\mathrm{d}n_{l_{i,j}}}{\mathrm{d}t} = F_{\mathrm{in}} x_{\mathrm{in}} + L_{j-1} x_{i,j-1} - V_j y^*_{i,j} + \mathbf{N}^\top r(\alpha) \quad (23)$$

Energy balance: Reactor stage

$$\begin{aligned}\frac{\mathrm{d}T}{\mathrm{d}t} =& \frac{1}{n_L C_{p_l}} \Big[F_{\mathrm{in}}(h_{\mathrm{in}} - h_{l_j}) + L_{j-1}(h_{l_{j-1}} - h_{l_j}) \\ & - V_j \Delta H_{\mathrm{vap},j} + \Delta H_r + UA(T_{\mathrm{jacket}} - T_j) \Big] \end{aligned} \quad (24)$$

$$\frac{\mathrm{d}T_{\mathrm{jack}}}{\mathrm{d}t} = \frac{1}{M_{\mathrm{oil}} C_{p_{\mathrm{oil}}}} \Big[F_{\mathrm{oil}}(h_{\mathrm{oil}} - h_{l_j}) + UA(T_j - T_{\mathrm{jack}}) \Big] \quad (25)$$

Pressure: Thermodynamic equilibrium

$$P_j = \sum_{i=1}^{S} x_{i,j} \gamma_{i,j}(x_{i,j}, T_j) P_j^{\mathrm{sat}}(T_j) \quad (26)$$

Reaction kinetics

$$r(\alpha) = K_0 \exp\left(\frac{-\alpha}{RT}\right) \prod_{i=1}^{S} [C]^{\nu_i} \quad (27)$$

where $n_l \in \mathbb{R}^S$ is the vector of moles in the liquid phase, $F_{in} \in \mathbb{R}^\iota$ is the inlet flow with composition x_{in}. y^* is the gas composition in equilibrium with the liquid bulk composition x. $\mathbf{N} \in \mathbb{R}^{R \times S}$ is the stoichiometric matrix, and r the vector of chemical reactions. T is the temperature, h_l is the liquid phase enthalpy, h_{in} is the enthalpy of the inlet stream, ΔH_r is the heat of reaction, ΔH_{vap} is the heat of vaporization, C_{p_l} is the heat capacity of the liquid phase, U is the overall heat transfer coefficient, A is the reactor surface area, M_{oil} is the total oil mass in the jacket, $C_{p_{\mathrm{oil}}}$ is the oil heat capacity, h_{oil} is the enthalpy of the oil at the inlet and T_{jack} is the jacket temperature. P_j is the absolute pressure, γ is the liquid activity coefficient, P^{sat} is the saturation pressure. α is the activation energy, $[C]$ is the concentration of the reactants, and ν is the stoichiometric coefficient. The parameters considered for analysis are $\mathbf{p} = [\alpha_1 \;\; \alpha_2 \;\; \alpha_3 \;\; \alpha_4 \;\; \cdots \;\; K_{0,1} \;\; K_{0,2} \;\; K_{0,3} \;\; K_{0,4} \;\; C_{p_{\mathrm{oil}}} \;\; U]^\top$. The nominal values for these parameters are shown in Table 1.

Table 1. Nominal values of selected parameters

Parameter	Value	Unit
Activation Energy 1 (α_1)	1.393272×10^5	kJ kmol^{-1}
Activation Energy 2 (α_2)	1.393272×10^5	kJ kmol^{-1}
Activation Energy 3 (α_3)	1.2675×10^5	kJ kmol^{-1}
Activation Energy 4 (α_4)	1.233×10^5	kJ kmol^{-1}
Pre-exp. factor 1 ($K_{0,1}$)	4.56×10^5	kg kmol^{-1} s^{-1}
Pre-exp. factor 2 ($K_{0,2}$)	9.7417×10^5	kg kmol^{-1} s^{-1}
Pre-exp. factor 3 ($K_{0,3}$)	5.73×10^8	kg kmol^{-1} s^{-1}
Pre-exp. factor 4 ($K_{0,4}$)	1.16939×10^9	kg kmol^{-1} s^{-1}
Oil heat capacity ($C_{p_{\mathrm{oil}}}$)	2.3	kJ kg^{-1} K^{-1}
Heat transfer coefficient (U)	1.8	kW m^{-2} K^{-1}

For this analysis, we perform an exploration up to 10% with respect to their nominal values in the positive and negative directions. In order to capture possible unknown nonlinear effects, the range of each parameter is partitioned using a centered linear spacing. Therefore,

$$T^5 = \{\mathbf{I}_{10}, -\mathbf{I}_{10}\}, \;\; M = \{0.02,\; 0.06,\; 0.1\} \times \mathbf{p}_{\mathrm{nom}}$$

where $\mathbf{p}_{\mathrm{nom}}$ correspond to the vector of parameters at their nominal values. The analysis is carried out assuming that each parameter remains constant at their adjusted value during each iteration. The measured variables in the process (outputs) are the reactor temperature T_r, the operating pressure in the reactor P_r, and the polymer viscosity μ. Furthermore, the variations of the outputs to the input signals are not considered because the process is already in closed-loop, and the input-output pairing has already been established by the process recipe. Additionally, Each of the EOSCM is normalized using the nominal parameter values and nominal trajectories to avoid that the units of parameters and outputs affect the sensitivity indexes.

As it can be seen in Figure 2, the most sensitive parameter is α_3. This parameter exhibits the largest trace (average total variation), the highest determinant (smallest correlation in the outputs), and largest inverse condition number (covariance matrix is well-conditioned). However, in this example it is clear that the use of a particular measure to rank parameters produces a substantial change. For example, the parameters $K_{0,3}$ and $K_{0,4}$ exhibit a large total average sensitivity, but inspecting the other indexes, it is clear that they have a large degree of correlation which

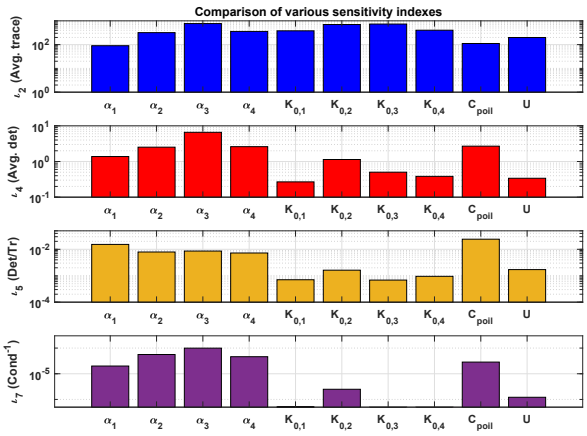

Fig. 2. Sensitivity values of the EOSCM evaluated for different sensitivity indexes.

makes them practically unidentifiable. Another interesting example is C_{poil}, this parameter is ranked very low with respect to the average trace, but based on the determinant and the inverse condition number, the oil heat capacity induces low correlation in the outputs, and generates a well-conditioned matrix covariance matrix. Furthermore, it is ranked first in terms of the trade-off between the average variation (average trace) and average correlation (average determinant), implying that this parameter can be estimated with low variance and correlation.

It must also be noted that by selecting an index to assess identifiability, an important aspect of the analysis is lost: the relative importance of a specific output for parameter estimation. For example the normalized EOSCM with respect to the highest ranked parameter α_3, which reads

$$\bar{W}_{C^o}[\alpha_3] = \begin{bmatrix} 0.0018 & -2.16\times10^{-5} & 0.058 \\ -2.16\times10^{-5} & 1.596\times10^{-6} & -0.000114 \\ 0.058 & -0.000114 & 1528.45 \end{bmatrix} \quad (28)$$

Inspecting the matrix in (28), it can be seen that the most important output to compute a reliable estimate for α_3 is μ, which exhibits both high sensitivity as well as low correlation with respect to the other outputs (T_r and P_r). This shows that parameter identifiability and output importance are a multidimensional problem that cannot be dealt with separately or using one index alone.

6. CONCLUSIONS

In this work, we have established the connection between sensitivity analysis and identifiability for large-scale nonlinear models using an output controllability approach and empirical covariance matrices. This representation allows us to address the identifiability of complex models from a practical point of view, while also explore larger parameter ranges and possible nonlinear effects. The approach is also extended to systems with no steady-states. The information contained in the empirical output sensitivity covariance matrix is similar to that of the observability matrix, but it also allows for the assessment of parameter sensitivity to input variation. This is advantageous for input design to maximize or minimize parametric sensitivity. Furthermore, since identifiability and sensitivity analyses using empirical covariance matrices are numerical approaches, these usually suffer from scaling issues. This is taken care of first by normalizing the empirical output sensitivity covariance matrix with nominal trajectories, and second, by choosing specific sensitivity indexes that could represent the parameter contribution to each available output. The approach has been implemented in a reactive batch distillation process to assess the identifiability of 10 parameters, where it is shown that indexes only provide limited information, and multiple indexes must be implemented to get sound conclusions. The empirical covariance matrix approach shows that four or five parameters are practically identifiable with respect to the specific operating trajectory.

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