OPERABILITY-BASED APPROACH FOR PROCESS DESIGN, INTENSIFICATION, AND CONTROL: APPLICATION TO HIGH-DIMENSIONAL AND NONLINEAR MEMBRANE REACTORS

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Abstract

Process operability was originally proposed as an approach for the design and control of complex chemical processes. This approach has been applied to processes that are described by high-dimensional linear models. For nonlinear systems, an operability-based method for design and intensification of low-dimensional membrane reactor systems has also been reported. However, operability algorithms for nonlinear systems are currently limited by the problem size that they can address. In this paper, an operability-based approach for process design, intensification and control is proposed to bridge this gap in the literature by facing the challenges of process nonlinearity and model size. The proposed framework relies on the combination of classical operability concepts with nonlinear programming tools for the computation of input and output sets in high dimensions. This study opens new avenues for process intensification research connected to design and control of advanced energy systems. To illustrate the effectiveness of the developed method, a membrane reactor system for the direct methane aromatization conversion to benzene and hydrogen is addressed as a shale/natural gas utilization example. The obtained results indicate that the proposed approach can be employed as a tool for input-output analysis of complex chemical and energy systems. These results also point out computational challenges associated with the design problem in high dimensions.

Keywords

Operability, Design, Intensification, Control, Optimization, Membrane reactor

Introduction and Prior Work

In recent years, due to the advent of the shale gas revolution, the US has incremented the annual production of processed natural gas by approximately 21% from 16.3 to 19.8 trillion cubic feet (EIA, 2016). These abundant amounts of shale gas will demand the development of tools for the techno-economic viability analysis of novel natural gas utilization processes that produce heat, power, fuels and chemicals. However, the design and control of such processes is a challenging task as these systems are typically operated in a highly constrained and integrated environment that is represented by complex large-scale and nonlinear models. In this paper, an operability-based approach is formulated for design, intensification and control purposes. The proposed method is applied to a membrane reactor system for the direct methane aromatization conversion to fuels and chemicals. This approach broadens the scope of the traditional path of operability methods for design and control, mainly focused on obtaining the achievable output set (AOS) from the available input set (AIS), to also allow the computation of the desired input set (DIS) from the desired output set (DOS), considering process constraints and intensification targets.

Process operability emerged in the last decades as a powerful tool for the design and control of industrial-scale chemical processes, including a variety of academic and industrial-scale applications from companies such as Air Products and Chemicals (APCI) and DuPont (Lima et al., 2010; Georgakis et al., 2003). In particular, operability enables the systematic verification of a design's ability to achieve the feasible region of operation for control in the presence of disturbances and provides a comparative measure to rank competing system designs (Vinson and Georgakis, 2000). Several systems studies that addressed the development and implementation of operability approaches are available in the literature. Specifically for high-dimensional and nonlinear systems, operability calculations using a response surface model-based approach have been performed (Georgakis and Li, 2010). In this work, the authors pointed out the computational limitations of the method for high-dimensional applications due to the

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size of the polynomial model typically used. To address this challenge, the authors proposed the selection of a finite number of points that provide an adequately accurate representation of the system. Optimization-based approaches to enable operability and flexibility studies of nonlinear systems have also been addressed. In particular, algorithms were developed for flexibility and optimization under uncertainty (Dominguez and Pistikopoulos, 2013; Liu et al., 2009). Refer to Lima et al. (2010a) and Georgakis et al. (2003) for an overview of the past operability and flexibility efforts, including systems of different dimensionality and nature. In more recent operability studies, approaches for design and intensification purposes have also been proposed focusing on low-dimensional membrane reactor (MR) systems for the direct methane aromatization (DMA) conversion to hydrogen and benzene (Carrasco and Lima, 2015; Carrasco and Lima, 2016). However, the reported literature review indicates lack of operability analysis for nonlinear high-dimensional energy systems. To bridge this gap, the objective in this manuscript is the development of algorithms based on classical operability concepts and nonlinear programming (NLP) tools for analyzing operating regions for process design, intensification, and control purposes. This manuscript is structured as follows. First, the operability concepts and a DMA-MR motivating example are presented. Then, the proposed operability-based approach is formulated for high-D systems. Finally, operability case studies for the DMA-MR system are performed focusing on the relationship between the input and the output sets.

Background

Operability Concepts

To perform the operability analysis for process design, intensification and control, the process model that describes the relationship between the input (manipulated and/or disturbance) and output variables needs to be known (Carrasco and Lima, 2015; Carrasco and Lima, 2016). This model ($\mathbf{M}$) can be represented by:

$$\mathbf{M} = \begin{cases} \dot{\mathbf{x}} = \mathbf{f}(\mathbf{x}, \mathbf{u}, \mathbf{d}) \\ \mathbf{y} = \mathbf{g}(\mathbf{x}, \mathbf{u}, \mathbf{d}) \\ \mathbf{h_1}(\dot{\mathbf{x}}, \mathbf{x}, \mathbf{y}, \dot{\mathbf{u}}, \mathbf{u}, \mathbf{d}) = \mathbf{0} \\ \mathbf{h_2}(\dot{\mathbf{x}}, \mathbf{x}, \mathbf{y}, \dot{\mathbf{u}}, \mathbf{u}, \mathbf{d}) \geq \mathbf{0} \end{cases} \quad (1)$$

in which $\mathbf{x} \in \boldsymbol{R}^r$ are the state variables of the process. Also, $\mathbf{u} \in \boldsymbol{R}^m$, $\mathbf{d} \in \boldsymbol{R}^q$ and $\mathbf{y} \in \boldsymbol{R}^n$ are the vectors of the manipulated inputs, the disturbances and the output variables, respectively. The time derivatives of $\mathbf{x}$ and $\mathbf{u}$ are denoted by $\dot{\mathbf{x}}$ and $\dot{\mathbf{u}}$, respectively, and $\mathbf{f}$ and $\mathbf{g}$ represent the nonlinear maps. The functions $\mathbf{h_1}$ and $\mathbf{h_2}$ include the process constraints associated with production and safety specifications. Finally, the model structure $\mathbf{M}$ can be defined by first principles, or process simulator runs can be used for obtaining the relationships between input and output spaces.

In particular, the manipulated inputs, $\mathbf{u}$, can take values in the available input set (AIS), based on the design of the process that is limited by the process constraints. Additionally, the disturbance variables, $\mathbf{d}$, which can represent the process uncertainties and variabilities, present values inside the expected disturbance set (EDS). Based on the process model, the achievable output set (AOS) for a specific disturbance vector, AOS($\mathbf{d}$), is defined by the ranges of the outputs, $\mathbf{y}$, that can be achieved using the inputs inside the AIS. For some cases, the AOS does not satisfy desired production requirements, such as production rates or product qualities. These requirements associated with the outputs would comprise the desired output set (DOS). These aforementioned sets can be mathematically represented as follows:

$$\mathbf{AIS} = \{\mathbf{u} \mid \mathbf{u}_i^{\min} \leq \mathbf{u}_i \leq \mathbf{u}_i^{\max}; \mathbf{1} \leq \boldsymbol{i} \leq \boldsymbol{m}\};$$
$$\mathbf{EDS} = \{\mathbf{d} \mid \mathbf{d}_i^{\min} \leq \mathbf{d}_i \leq \mathbf{d}_i^{\max}; \mathbf{1} \leq \boldsymbol{i} \leq \boldsymbol{q}\}$$
$$\mathbf{AOS}(\mathbf{d}) = \{\mathbf{y} \mid \mathbf{y} = \mathbf{M}(\mathbf{u}, \mathbf{d}); \mathbf{u} \in \mathbf{AIS}, \mathbf{d} \text{ is fixed}\};$$
$$\mathbf{DOS} = \{\mathbf{y} \mid \mathbf{y}_i^{\min} \leq \mathbf{y}_i \leq \mathbf{y}_i^{\max}; \mathbf{1} \leq \boldsymbol{i} \leq \boldsymbol{n}\}$$

Finally, considering that the DOS can be defined as the output space where the process should be operated, the set of inputs required to reach the entire DOS for a specified $\mathbf{d}$ vector is defined as the desired input set, DIS($\mathbf{d}$). In particular, if the DOS is a subset of the AOS, the calculated DIS($\mathbf{d}$) can be obtained by model inversion, as shown below:

$$\mathbf{DIS}(\mathbf{d}) = \{\mathbf{u} \mid \mathbf{u} = \mathbf{M}^{-1}(\mathbf{y}, \mathbf{d}); \mathbf{y} \in \mathbf{DOS}, \mathbf{d} \text{ is fixed}\}$$

Membrane Reactor Motivating Example

In the last decades, membrane reactors (MRs) have emerged as examples of intensified processes as they combine reaction and separation unit operations in one process improving the system efficiency, lowering the cost and minimizing the environmental footprint (Drioli et al., 2011). MRs also allow higher conversions than conventional packed-bed reactors due to the reaction equilibrium shift towards the products caused by the selective removal of one of these products through the membrane. Specifically, in the DMA process for production of H_2 and C_6H_6, the reaction equilibrium is shifted towards products as H_2 is removed through a H_2-selective membrane (ion transport-based). Restrictions on process target specifications, such as reaction conversions and production rates, as well as multiple challenges associated with controlling temperatures and compositions, make the problem of designing and

controlling MRs for the DMA process a challenging research problem. Motivated by these challenges, the DMA-MR example is selected here to illustrate the nonlinear operability concepts. Fig. 1 shows a schematic representation of the MR considering a cocurrent flow configuration. The following 2-step reaction mechanism is assumed for this process:

Step 1: *Step* 2:

$$2\,CH_4 = C_2H_4 + 2\,H_2 \qquad 3\,C_2H_4 = C_6H_6 + 3\,H_2$$

$$r_1 = k_1 C_{CH_4}\left(1 - \frac{k_1' C_{C_2H_4} C_{H_2}^2}{k_1 C_{CH_4}^2}\right) \qquad r_2 = k_2 C_{C_2H_4}\left(1 - \frac{k_2' C_{C_6H_6} C_{H_2}^3}{k_2 C_{C_2H_4}^3}\right)$$

in which C_{CH_4}, $C_{C_2H_4}$, C_{H_2}, and $C_{C_6H_6}$ are the concentrations in the gas phase of methane, ethene, hydrogen, and benzene, respectively. Also, r_1 and r_2 represent the reaction rates of steps 1 and 2, respectively, and k_1, k_1', k_2, and k_2' correspond to the reaction rate constants (where the symbol ´ denotes the inverse reaction).

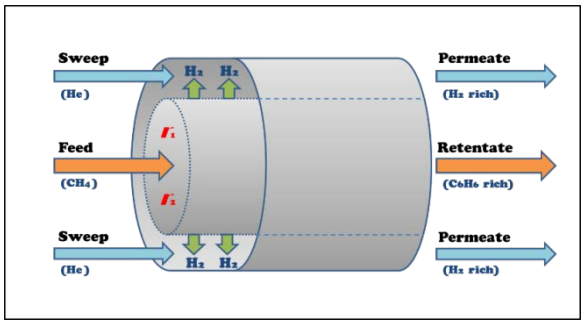

Figure 1. Schematic of DMA-MR with a cocurrent configuration

Considering the shell and tube MR design in Fig. 1, in which CH_4 feeds the reaction side (tube packed with catalysts) and the sweep gas (e.g., helium) flows in the permeation side (shell), the H_2 produced in the tube permeates to the shell through the membrane layer that is placed on the surface of the tube wall. The produced outlet streams from the tube (retentate) and shell (permeate) are rich in C_6H_6 and H_2, respectively (see Carrasco and Lima, 2015 and 2016, for details on the model derivation, including mass balances, the derived ordinary differential equations (ODE) and simulation conditions).

Table 1. Input-output ranges that define the AIS and AOS

Input variables	**Available ranges**
Reactor length [cm]	10 – 100
Tube diameter [cm]	0.5 – 2.0
Permeance [mol/(s.m^2.atm$^{1/4}$)]	1x10^{-4} -0.01
Selectivity [-]	100 – 1x10^5
Output variables	**Achievable ranges**
Benzene production [mg/h]	0 - 26
Hydrogen production [mg/h]	0 – 7
Methane conversion [%]	10 – 46
Cost factor [-]	100 - 1,000

Operability Analysis for Computation of Achievable Regions for Control

The MR operability analysis will be carried out by mapping the feasible regions between process input and output variables. For this case study, a 4x4 system is initially analyzed. Table 1 shows the four selected input/design variables and their available ranges in the AIS. This table also shows the four output variables with their achievable ranges in the AOS, obtained by the operability input-output mapping. Although the disturbance variable, **d**, is considered in the proposed method, for this application, the value of **d**=0 is assumed (operating point without disturbances).

For the performed operability analysis, a grid is built for the input variables by assuming that their ranges are equally divided by 5, thus 625 points are evaluated. The hydrogen production rates are calculated from the permeate side, and a cost factor for comparing different designs is considered, assuming estimated cost parameters associated with the membrane area, the catalyst mass, and the reactor volume. Table 2 presents the details for the calculated output ranges including some suggested zones for further analysis.

Table 2. Achievable ranges for output variables

Output variables	**Achievable ranges and zones**		
	a	**b**	**c**
Benzene production [mg/h]	10 - 15	20 - 26	0 - 10
Hydrogen production [mg/h]	0 - 4	2 - 4	4 - 7
Methane conversion [%]	10 - 30	40 - 45	40 - 46
Cost factor [-]	100 - 300	100 - 1000	100 - 1000

The achievable output ranges are also shown in Fig. 2 as a 4-D plot, in which the plotted points are color coded with respect to their cost values and some of the 5^4 points considered are superposed. These ranges could help defining achievable set points or ranges for an advanced controller (e.g., MPC). Regarding the specific zones in this figure, zone “b” represents the best balance between benzene and hydrogen production rates with a high methane conversion, in a wide range of costs. According to the operability analysis, this zone is obtained when the permeance and selectivity are at their highest values, 0.01 [mol/(s.m^2.atm$^{1/4}$)] and 100,000, respectively. This performance level can be explained by the smaller permeation of benzene through the membrane at these conditions, for also small membrane surface areas. For instance, for the mentioned membrane specifications and a tube length and diameter of 10 [cm] and 2 [cm], respectively, it is possible to obtain

production rates of benzene and hydrogen, a methane conversion, and a cost factor of approx. 25.4 [mg/h], 3.9 [mg/h], 40.3 [%], and 110, respectively. If the tube length is changed to 100 [cm], the calculated production rates of benzene and hydrogen, methane conversion and cost factor reach values of about 24.8 [mg/h], 3.9 [mg/h], 40.4 [%], and 1,000, respectively. These results indicate that similar membrane reactor performance levels can be obtained for different footprints (approx. 90% difference in catalyst volume and membrane surface area between the two cases) and thus cost values. Also, with the operability analysis performed, it is possible to identify that the maximum hydrogen production rate in zone "b" is ≈4 [mg/h], which could be increased up to 7 [mg/h] in other zones. In contrast, zone "c" shows the lowest benzene production rate caused mainly by low membrane selectivity and high permeance, for reactor length and diameter tending to their high limits. For example, when the membrane permeance, selectivity, tube length and diameter are 0.01 [mol/(s.m^2.atm$^{1/4}$)], 100, 100 [cm] and 2 [cm], respectively, the results show undesired performance in terms of production rate of benzene (almost zero), for a high methane conversion value of 45.7 [%]. This outcome suggests that high methane conversions do not necessarily translate to good levels of benzene production rates.

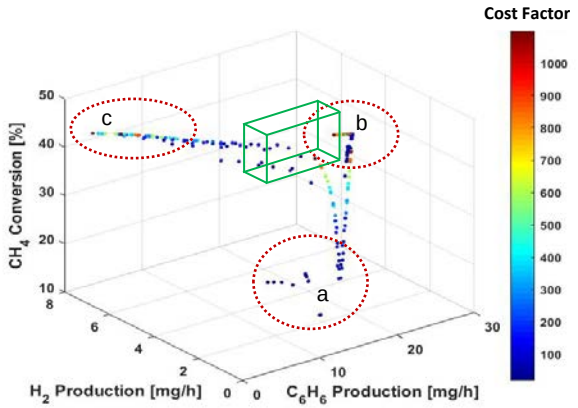

Figure 2. 4-D AOS and DOS for DMA-MR Example

Finally, the low methane conversion values for zone "a" occur due to the low membrane permeance considered of 1x10^{-4} [mol/(s.m^2.atm$^{1/4}$)]. In this case, the reaction equilibrium shift towards products is limited due to the low H_2 removal through the H_2-selective membrane.

The computational time required to perform the operability analysis for the 4x4 system was 102 seconds as presented in Table 3. This table also shows additional information regarding the computational times for different subsystems, 2x2 (inputs: reactor length and diameter; outputs: methane conversion and benzene production) and 3x3 (inputs: reactor length and diameter, and membrane selectivity; outputs: methane conversion, benzene and hydrogen productions), of the DMA-MR for comparison. These results show that the computation of achievable output regions for control purposes could be completed in order of minutes or even seconds depending on the considered system dimensionality. All the operability calculations in this paper were carried out on an Intel Core i7 (Sandy bridge) 3.40 GHz processor.

Table 3. Computational time for AOS calculations for different DMA-MR subsystems

System	Points	Time [min:sec]
2x2	25	00:03
3x3	125	00:18
4x4	625	01:42

Now, for design and intensification purposes, if the DOS (depicted in Fig. 2 as a green parallelepiped) is not a subset of the AOS, scenario that would likely occur in practice, the feasible DIS can be calculated to provide design changes that must be performed. For this calculation, an optimization algorithm based on NLP tools is formulated next, considering process constraints and target specifications.

Optimization-based Operability Approach for Design and Intensification

Problem Formulation

Problem P1 addresses the minimization of the objective function Φ, mathematically defined by the relative error (or distance) between the desired output points from the DOS ($y_{j,k}$) and the feasible output points ($y^*_{j,k}$), as follows:

$$\text{P1:} \qquad \Phi_k = \min_{u_k^*} \sum_{j=1}^{n} \left((y_{j,k} - y^*_{j,k}) / y_{j,k} \right)^2$$

subject to: nonlinear model (eq. 1)

$$u_k^{min} \le u_k^* \le u_k^{max}$$

$$\mathbf{c}_1(u_k^*) \le \mathbf{0}$$

in which $y_{j,k}$ corresponds to the DOS selected points according to a specified grid. The subscript *j=1,2,3…n* represents the output variable index, and *k=1,2,3…p* is defined as the point index for the total points in a grid. The selected grid depends on the level of DOS discretization. Thus, the DOS can also be represented as

$$\text{DOS} = \{ \mathbf{y} \mid \mathbf{y} = (y_1, y_2, y_3 \ldots y_p) \}$$

in which, the desired output points are elements of the output set, $y_k = (y_{1,k}, y_{2,k} \ldots y_{j,k} \ldots y_{n,k})$.

The result from the optimizer in P1 consists of the input set calculated from the different points, u_k^*, that defines the DIS* as the feasible DIS that is given by

$$\mathrm{DIS}^* = \{\mathbf{u}^* \mid \mathbf{u}^* = \left(u_1^*, u_2^*, u_3^* \ldots u_p^*\right)\}$$

in which, the feasible desired input points can be written as $u_k^* = \left(u_{1,k}^*, u_{2,k}^* \ldots u_{i,k}^* \ldots u_{m,k}^*\right)$ where the subscript *i=1,2,3…m* denotes the input variable index. The feasible DOS, DOS*, is formed by all the calculated $y_{j,k}^*$, points that are the closest to their counterparts ($y_{j,k}$) in terms of distance, according to the objective function defined in P1. The DOS* can also be calculated using the DIS* and the process model, **M**, by direct mapping as follows:

$$\mathrm{DOS}^* = \{\mathbf{y}^* | \mathbf{y}^* = \mathbf{M}(\mathbf{u}^*); \mathbf{u}^* \in \mathrm{DIS}^*\}$$

To start the optimizer evaluations, an initial guess input point, $u_{k,o}^*$, that is within the AIS ranges is requested. Finally, $\mathbf{c}_1$ represents the nonlinear constraint map associated with specified design conditions. Thus, the optimization problem P1 is characterized by a nonlinear objective function, with nonlinear constraints, which can be solved by the NLP solvers available in the Optimization toolbox in MATLAB (in this case "fminsearch").

DMA-MR Example Revisited

In order to include desired output requirements in the operability analysis, in this case benzene and hydrogen production rates, as well as methane conversion and cost, the DOS should now be specified as shown in Table 4. Considering that zone "b" presented a good balance among the output variables as discussed above (see Fig. 2 and Table 2), the DOS is defined around this zone. In particular, it is desirable to push the production rate of hydrogen as much as possible from the original 2 – 4 [mg/h] to 4 – 5 [mg/h] for a reduced cost factor range between 100 - 400. Thus, the desired production levels for hydrogen are outside the original achievable range calculated from the AIS. To provide the feasible operating envelope for the DIS* given the constraints specified by the DOS, the optimization-based operability algorithm defined in P1 is employed. Table 4 also presents the ranges calculated for the DIS* and DOS*as outcomes of P1.

For this example, the DOS is discretized considering *p* = 625 points (i.e., the domains for the benzene and hydrogen production rates, methane conversion, and cost factor are divided in a 5x5x5x5 grid), thus *k=1,2,3…625*. For the DIS* calculation, design constraints associated with the plug flow reactor assumption, such as length over diameter ≥ 30, reactor length for an experimental setup less than 300 [cm], and membrane permeance and selectivity less than 0.01 [mol/(s.m^2.atm$^{1/4}$)] and 10,000, respectively, are also incorporated into the problem as constraints.

Table 4. Specified DOS, calculated DIS and DOS* ranges*

Desired output variables	**Desired output ranges (DOS)**
Benzene production [mg/h]	15 - 25
Hydrogen production [mg/h]	4 – 5
Methane conversion [%]	35 - 45
Cost factor [-]	100 - 400
Feasible input variables	**Feasible input ranges (DIS*)**
Reactor length [cm]	26 – 300
Tube diameter [cm]	0.2 – 1.3
Permeance [mol/(s.m^2.atm$^{1/4}$)]	4x10^{-4} – 0.01
Selectivity [-]	72 – 1x10^4
Feasible output variables	**Feasible output ranges (DOS*)**
Benzene production [mg/h]	14 - 24
Hydrogen production [mg/h]	3.9 – 4.5
Methane conversion [%]	40 - 43
Cost factor [-]	100 - 400

Also, positivity restrictions on input variables are imposed. The optimization problem is solved for all of the DOS points in the established grid.

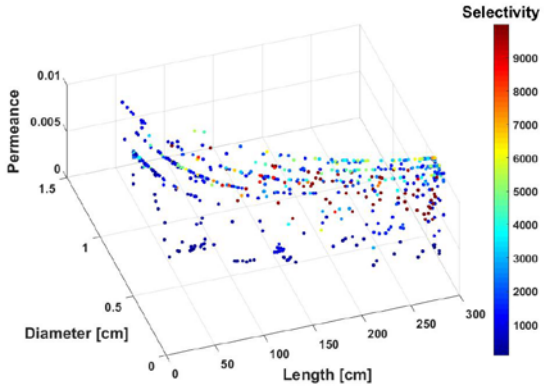

Figure 3. 4-D DIS for DMA-MR Example*

The DIS* outcome from the optimizer for all DOS points is shown in Fig. 3. Note that the calculated feasible ranges for the input variables are not completely contained in the initial AIS (see Table 1 for the AIS ranges). These results thus provide the input points that would be necessary to satisfy the DOS, in terms of process design specifications. Therefore, the space DIS* is comprised of the input points that satisfy the specified target specifications and constraints.

The calculated DOS* presented in Table 4 shows benzene production rate and cost factor ranges that satisfy their respective desired output ranges for the most part. Also, the range of the calculated hydrogen production rate is pushed from 2.0 – 4.0 [mg/h] to 3.9 – 4.5 [mg/h] showing an improvement with respect to the original range in zone "b" of the AOS (see Table 2). The obtained mapping thus can also provide the input-output points that fall within the desired ranges, for example in zone "b".

Fig. 4 depicts the zoomed versions of the DOS (see green parallelepiped) and the calculated DOS* (see solid dots). It also shows two zones "r" and "s", where the benzene and hydrogen production rates are less than 15 [mg/h] and 4 [mg/h], respectively.

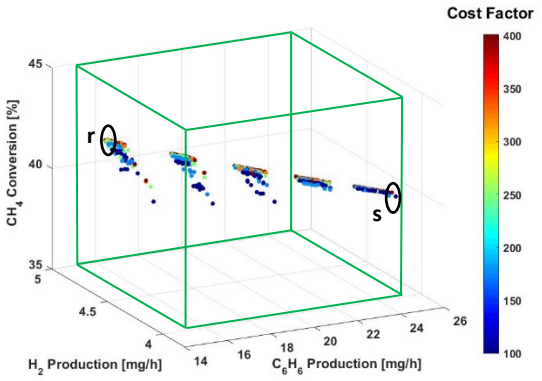

Figure 4. 4-D DOS and DOS for DMA-MR Example (Zoomed Versions)*

Table 5 presents the computational time required to perform the operability computations for the 4x4 system addressed. This table also shows information for 2x2 and 3x3 DMA-MR subsystems. The high computational time for the 4-D system should not be an issue if offline calculations were to be performed for design and intensification purposes.

Table 5. Computational time for DIS calculations for DMA-MR systems of different dimensionalities

System	Points	Time [hr:min:sec]
2x2	25	00:06:15
3x3	125	01:25:02
4x4	625	13:08:08

Conclusions

This article introduced a novel operability-based approach for process design, intensification and control of high-dimensional nonlinear systems. As an example of application of the proposed approach, a 4-D DMA-MR system was addressed. The obtained results showed the successful computation of achievable regions for control purposes. For the design case, the optimization-based operability calculations provided the necessary process design modifications to achieve feasible and desired output targets. The obtained results also showed that different reactor designs with similar levels of performance may have significantly different process footprints (up to 90%) in terms of reactor volume. This study opens new avenues for process intensification research connected to design and control of advanced energy systems. Our current research is focused on addressing the computational challenges associated with the nonlinear programming problem solved for design purposes to ultimately tackle a high-dimensional natural gas combined cycle process example. Future research will also focus on the implementation of a constrained controller to employ the computed achievable regions for the outputs.

Acknowledgments

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