A Reduced-order Model for Real-time NMPC of Ethanol Steam Reformers ⋆

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Abstract: One of the approaches to green energy of interest in recent years is through the use of hydrogen technologies in which the waste product of combustion is water rather than carbon dioxide, nitrous oxides, and other pollutants generated by the burning of hydrocarbon fuels. Ethanol steam reformers is one of the primary technologies with the potential to generate hydrogen efficiently and reliably. This article investigates the design of nonlinear optimal feedback control systems for ethanol steam reformers based on mechanistic models described by a system of nonlinear partial differential-algebraic equations, aka singular/descriptor systems. The mechanistic models are too complicated to simulate in real-time using the standard numerical discretization methods, much less to directly incorporate such models as nonlinear constraints into mechanistic model-based nonlinear model predictive control formulations. This article employs a rather sophisticated multivariate form of the method of characteristics to reformulate the nonlinear constraints into a much simpler form amenable to real-time control implementations.

Keywords: Modeling for control optimization, Descriptor systems, Model reduction of distributed parameter systems, Energy systems, Nonlinear model reduction, Model reduction

1. INTRODUCTION

Hydrogen technologies have the potential to significantly reduce air and water pollution and greenhouse emissions from automobiles compared to the hydrocarbon-based fuels of today, i.e., gasoline, jet fuel, natural gas, and ethanol. The waste product of using hydrogen in automobiles, when used in hydrogen fuel cells with oxygen, is water. A drawback of employing hydrogen as a fuel in automotive applications is that the hydrogen posed an explosion hazard associated with it having the highest gravimetric energy density (kJ/g) of all known substances, and that the hydrogen must be stored under very high pressure for the volume of the fuel tank to be practical. One of the promising technologies for providing safe hydrogen transport is to store the fuel in the form of a low-pressure liquid, and then use an on-board reformer to generate hydrogen when needed (Zhang et al., 2016). A highly selective membrane is used to allow only the hydrogen gas to exit the reformer to feed the fuel cell, with the other products of low energy density stored on-board until time to re-fuel. During refilling of the fuel tank, the products in a separate tank are pumped out of the vehicle and later processed in facilities at much higher efficiencies than what is feasible on the automotive applications. Of the reforming technologies, ethanol steam reforming is among the most promising.

The use of ethanol steam reformers for hydrogen production requires the design of real-time feedback control strategies to ensure efficient operation of the entire system while suppressing the effects of disturbances such as the large variations in internal and external temperatures during automotive operation. The limited control studies for ethanol steam reforming include decoupled proportional-integral-derivative (PID) controllers (Biset et al., 2009; García et al., 2010; Garcia et al., 2013) which do not explicitly take constraints into account, model predictive control systems which explicitly take constraints into account (e.g., (Recio-Garrido et al., 2012; Perez et al., 2016),(Serra et al., 2017)). These studies include the implementation of a real-time model predictive control algorithm based on a linear input-output model and so did not require estimator design (Torchio et al., 2016). Although a linear control algorithm will give adequate closed-loop performance for a small enough region of operation, as demonstrated in that study, the full range of potential operation induced much

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larger nonlinearities, and a nonlinear model predictive control algorithm would give better closed-loop performance – at least in the case in which input, state, and output constraints are explicitly taken into account.

As a first step towards nonlinear model predictive control design, we recently completed the first detailed theoretical analysis of the nonlinear dynamics of ethanol steam reforming (Reyero et al., 2020). By applying a null-space analysis, the mechanistic model was shown to be a singular (aka descriptor) distributed parameter system, namely, that the governing equations of energy and species material balances had both spatially distributed and algebraic character. Further, a method was proposed for the construction of approximate reduced-order nonlinear dynamics models that reduced the computational cost while retaining most of the structure of the original governing equations and the dependency on physical model parameters.

While the resulting reduced-order models could be directly incorporated into real-time nonlinear model predictive control, some assumptions were made that limited the practicality of the results. The assumptions were that the temperature was constant and the axial flow velocity had a weak dependency on the species concentrations and temperature. Both assumptions reduce the amount of nonlinearity in the model dynamics, and result in model uncertainty should the reduced-order model be incorporated into a nonlinear model predictive control algorithm. This article removes these assumptions by taking a much more sophisticated approach that introduces *zero error* in the model reformulation. This new formulation retains the same advantages of dependency on physical model parameters and greatly reducing the real-time computations required when incorporating the model into a nonlinear model predictive control algorithm. The theory has some novel elements particular to the steam reforming system, including the derivation of an analytical solution of the generalized eigenvalues and left generalized eigenvectors of an 8×8 matrix.

The remainder of this article is organized as follows. Section 2 summarizes the ethanol steam reformer system under consideration including the involved chemical reactions and the resultant nonlinear model. Section 3 describes a class of nonlinear distributed parameter systems – quasi-linear hyperbolic vector equations – which is a superset for the governing equations for ethanol steam reforming. Section 4 describes a mathematical derivation of the methods of characteristics that applies to the systems of first-order hyperbolic equations in Section 3. Section 5 employs these expressions with some additional theoretical analysis to derive a reduced-order model which has much lower computational cost, by exactly transforming the original system of eight nonlinear partial-differential equations to a systems of ordinary differential equations that provide precisely the state information needed in a nonlinear model predictive control design. Finally, Section 6 summarizes the main conclusions.

2. SYSTEM DESCRIPTION

This article considers an ethanol steam reformer (ESR) as a nonlinear dynamical system that consists of a catalytic

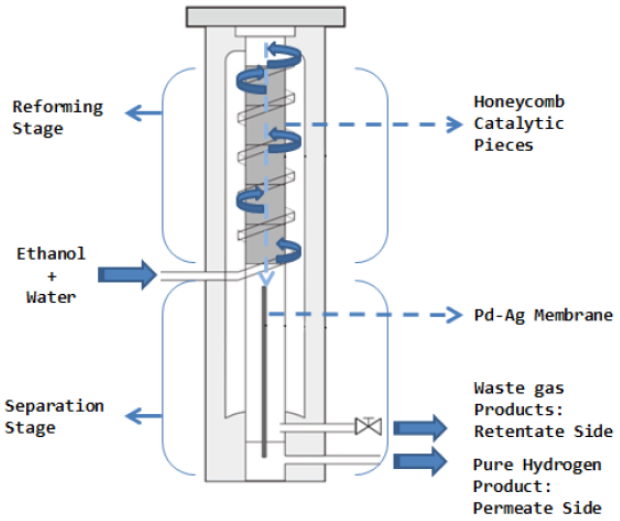

Fig. 1. Staged membrane reactor scheme

ethanol steam reactor in series with a a separation stage that incorporates a selective membrane for removal of the hydrogen (Perez et al., 2016; Reyero et al., 2020). The two unit operations are placed inside of a single integrated module, called a *staged membrane reactor* (López et al., 2012). Since the system is described in detail in the past publications, this section provides only a high-level summary.

The chemical reactions that occur in the ethanol steam reformer are (Uriz et al., 2011)

$$C_2H_5OH \longrightarrow CH_3CHO + H_2 \tag{1a}$$
$$C_2H_5OH \longrightarrow CO + CH_4 + H_2 \tag{1b}$$
$$CO + H_2O \rightleftharpoons CO_2 + H_2 \tag{1c}$$
$$CH_3CHO + 3\,H_2O \longrightarrow 2\,CO_2 + 5\,H_2 \tag{1d}$$

The chemical reactions occur in a tubular packed-bed reactor with a single inlet and a single outlet.

With modest assumptions detailed in past work (Reyero et al., 2020), the mechanistic model for the two process units are a function of the time and axial direction, that is, have only one spatial dimension in the partial differential equations describing the nonlinear dynamics of the system. The eight governing equations for each unit operation consist of seven species material balances and one energy balance, and have the same mathematical structure, which is in the form of a system of first-order quasi-linear hyperbolic equations described in the next section.

3. QUASI-LINEAR HYPERBOLIC EQUATIONS

Consider the class of nonlinear distributed parameter systems described by the first-order quasi-linear hyperbolic vector equation

$$A(x, y, u)u_x + B(x, y, u)u_y = g(x, y, u), \tag{2}$$

where $u(x, y) \in \mathbb{R}^m$ is an vector of distributed states with each element being a function of the real independent variables x and y, $A(x, y, u) \in \mathbb{R}^{m\times m}$ and $B(x, y, u) \in \mathbb{R}^{m\times m}$ are real matrices, $g(x, y, u) \in \mathbb{R}^m$ is a vector with

each element being an algebraic function of its arguments, and $u_x := \frac{\partial u}{\partial x}$ and $u_y := \frac{\partial u}{\partial y}$. Each individual equation in (2) arises from conservation of some property, such as mass, moles, particles, energy, or momentum.

The system (2) arises in numerous applications involving fluid flows, broadly speaking, for systems in which the overall dynamics are much more strongly affected by advection (i.e., bulk fluid flow) than diffusive phenomena (e.g., molecular diffusion, thermal diffusion, and viscosity). Whether this assumption is valid can be assessed by computing the Peclét number, which is the ratio of scalars that quantify the relative importance of advective to diffusive transport (Deen, 2011). For example, the Peclét number $\mathrm{Pe} = Lv/D$ for mass transfer and $\mathrm{Pe} = Lv/\alpha$ for heat transfer, where L is the relevant length scale (e.g., maximum dimension of the system), v is the local velocity, D is the molecular diffusivity, α is the thermal diffusivity.

In dynamic control applications, one of the independent variables in (2) is time and the other is a spatial coordinate or internal property such as particle size or age (in the latter case, the equation is typically called a population balance model, e.g., Gunawan et al. (2004); Rawlings et al. (1993); Nagy and Braatz (2012)). The above governing equation is a partial differential equation and said to be *regular* if the matrix corresponding to the time variable is nonsingular for all values of x, y, and u that can arise during the dynamics, and is a partial differential-algebraic equation and said to be *singular* (aka *descriptor*) if the matrix can be singular for some x, y, and u. In nearly all applications, the rank of the matrix is the same for all allowable values of all values of x, y, and u due to structural constraints induced by modeling assumptions. In other words, the structural rank of the matrix is equal to the rank of the matrix for any allowable value of x, y, and u. Although this article considers only two independent variables as that occurs in a large number of control applications, the overall approach and results generalize to systems with more than two independent variables in a straightforward manner.

Many optimal control algorithms have been developed for the case where (2) is regular, with the vast majority of that literature considering the case of $m = 1$. Optimal control design for the much more challenging case where (2) is singular and $m > 1$ has been investigated by the authors (Reyero et al., 2020), motivated by the specific application of optimal control of steam reforming. After exploring the models in some detail, we came to realize that the system class (2) was actually quite pervasive in real process control applications, including in tubular chemical reactors and chromatography columns ranging from the micro- to the macroscale. While the study and implementation of optimal control systems for tubular chemical reactors have a long history (Georgakis et al., 1977; Zavala and Biegler, 2009), we have not been able to locate any publications that formulate a nonlinear optimal feedback control system for singular systems described by (2) that is implementable with low online computational cost, i.e., that did not require some form of spatial discretization of the partial differential equations. The drawback of such formulations for automotive applications is that the incorporation of such models into nonlinear model predictive control algorithms results in a high computational cost that would be better used for moving the automotive vehicle than for carrying out the control calculations.

The main objective of this article is to present the first such mathematical formulation, which we write as a nonlinear model predictive control (NMPC) algorithm. The existing NMPC formulations are all too computationally expensive to be implementable in real time for $m \gg 1$, so a new formulation was derived.

The next section describes theoretical background on the method of characteristics for solving singular first-order quasi-linear hyperbolic vector equations.

4. METHOD OF CHARACTERISTICS

The method of characteristics is an approach for simplifying the computations associated with simulating hyperbolic partial differential equations, which has been used in applied mathematics for decades (Courant and Hilbert, 1962) but not as heavily exploited in optimal control theory and algorithms (some exceptions are Shang et al. (2004) and Munusamy et al. (2013)). There are several ways to interpret the method of characteristics, including as a change between moving and non-moving reference frames, as a complicated change in variables, as an exercise in differential algebra, or in terms of vector calculus.

This section describes the mathematical approach used to replace what are by far the most computationally expensive constraints in a nonlinear model predictive control formulation, which are partial differential-algebraic equations, with system of ordinary differential equations that produce exactly the state information needed by the control algorithm. The mathematical analysis in this section draws heavily on the theory on the numerical analysis and simulation of hyperbolic systems of quasilinear systems, especially the book (Rhee et al., 2001). To simplify the form of the expressions, the arguments are not written explicitly below.

The first step is to multiply both sides of (2) by a row vector $\boldsymbol{l}^\top$ with $\boldsymbol{l} \in \mathbb{R}^m$:

$$\boldsymbol{l}^\top A u_x + \boldsymbol{l}^\top B u_y = \boldsymbol{l}^\top g. \tag{3}$$

The method of characteristics analyzes the dynamics in terms of a *characteristic curve*, which is the ordered pair $(x(\omega), y(\omega))$ parameterized by the real scalar ω. Multiplying each term on the left-hand side of (3) by $x(\omega)$ and $y(\omega)$ gives

$$\left(\frac{1}{x_\omega}\boldsymbol{l}^\top A\right) u_x x_\omega + \left(\frac{1}{y_\omega}\boldsymbol{l}^\top B\right) u_y y_\omega = \boldsymbol{l}^\top g. \tag{4}$$

Now if a vector $\boldsymbol{l}$ and ordered pair $(x(\omega), y(\omega))$ can be found such that

$$\frac{1}{x_\omega}\boldsymbol{l}^\top A = \frac{1}{y_\omega}\boldsymbol{l}^\top B, \tag{5}$$

then (4) can be written as

$$\left(\frac{1}{x_\omega}\boldsymbol{l}^\top A\right)(u_x x_\omega + u_y y_\omega) = \boldsymbol{l}^\top g. \tag{6}$$

Application of the chain rule

$$u_\omega = u_x x_\omega + u_y y_\omega \tag{7}$$

and multiplication by a scalar simplifies this expression to

$$\boldsymbol{l}^\top A u_\omega = \boldsymbol{l}^\top g x_\omega. \tag{8}$$

Application of (5) gives the equivalent expression

$$\boldsymbol{l}^\top B u_\omega = \boldsymbol{l}^\top g y_\omega. \tag{9}$$

The above expressions can be written using the chain rule as

$$\boldsymbol{l}^\top A \frac{du}{dx} = \boldsymbol{l}^\top g \tag{10}$$

and

$$\boldsymbol{l}^\top B \frac{du}{dy} = \boldsymbol{l}^\top g. \tag{11}$$

The next step is to characterize the set of vectors $\boldsymbol{l}$ and ordered pairs $(x(\omega), y(\omega))$ that satisfy (5). The expression (5) defines the left generalized eigenvalue decomposition, that is, multiplication by x_ω gives the equivalent expression

$$\boldsymbol{l}^\top (A - \sigma B) = 0. \tag{12}$$

where

$$\sigma = \frac{x_\omega}{y_\omega} = \frac{dx}{dy}. \tag{13}$$

Equation (12) has m solutions (possibly repeated) defined by the generalized eigenvalues σ_k and left generalized eigenvectors $\boldsymbol{l}_k$:

$$\boldsymbol{l}_k^\top (A - \sigma_k B) = 0, \quad k = 1, 2, \ldots, m. \tag{14}$$

Subroutines for computing the generalized eigenvalues and eigenvectors given A and B – which are known matrices for any values of x, y, and u – are available in most linear algebra-related software packages including in LAPACK and Matlab (the *eig* command). Many algorithms are available, including the QZ algorithm which requires $\mathcal{O}(n^3)$ floating point operations and $\mathcal{O}(n^2)$ memory locations (Moler and Stewart, 1973; Demmel, 1997; Golub and Loan, 1996) and is an extension of the QR algorithm commonly used for regular eigenvalue problems. In applications, A, B, or both are sparse, and iterative solvers have been derived that exploit sparsity to reduce computations and memory and handle very high dimensionality (e.g., see discussions in (Hao et al., 2016)).

Collecting (8) and (13) into a system of ODEs,

$$\begin{aligned} \frac{\mathrm{d}y}{\mathrm{d}x} &= \frac{1}{\sigma_k(x,y,u)}, \\ \boldsymbol{l}_k(x,y,u)^\top A(x,y,u) \frac{\mathrm{d}u}{\mathrm{d}x} &= \boldsymbol{l}_k(x,y,u)^\top g(x,y,u), \\ & \qquad k = 1, 2, \ldots, m. \end{aligned} \tag{15}$$

This system of $2m$ equations can be solved using any standard ODE solver such as forward Euler-type numerical scheme (Rhee et al., 2001). The above equations are solved with an initialization being the state at the inlet boundary condition for the non-time coordinate, and then collected together to form ordered triplets (x, y, u), which are values of the vector u for each ordered pair (x, y).

The characteristic equations can be integrated with respect to time for any specific inlet conditions, and the resulting solutions can be combined to construct all of the distributed states, namely, temperature and species concentrations. The complexity of the model equations that serve as constraints in a nonlinear model predictive control formulations are greatly reduced by replacing the distributed parameter model (2) by a system of ODEs. This reduced-order control-relevant model can be directly incorporated into any standard nonlinear MPC formulation.

5. REDUCED-ORDER MODEL FOR ETHANOL STEAM REFORMING

This section applies the above expressions for the general first-order quasi-linear hyperbolic equation (2) to the specific model for ethanol steam reforming given by Reyero et al. (2020).

The model equations can be written as (Reyero et al., 2020)

$$M f_t + N f_z = g(t, z, f), \tag{16}$$

with [1]

$$f := \begin{pmatrix} F \\ T \end{pmatrix}, \quad F = \begin{pmatrix} F_1 \\ \vdots \\ F_7 \end{pmatrix}, \tag{17}$$

$$M = \begin{pmatrix} I - \dfrac{1}{\|F\|} F e^\top & -\dfrac{1}{T} F \\ 0^\top & \dfrac{C_v}{RT\|F\|} F \end{pmatrix}, \tag{18}$$

$$N = \begin{pmatrix} \dfrac{RT}{Ap} I & 0 \\ \dfrac{RT}{Ap} e^\top & \dfrac{C_p}{Ap} F \end{pmatrix}, \tag{19}$$

where

- I is the 7×7 identity matrix,
- $e \in \mathbb{R}^7$ is the vector of ones,
- F_j is the flow rate of species j,
- $\|F\|$ is the Euclidean norm of the vector F,
- T is the temperature (in K),
- R is the ideal gas constant (in $\mathrm{Pa\,m^3/(mol\,K)}$),
- ρ is the membrane thickness (in m),
- c_p and c_v are the heat capacities (in J/(mol K)).
- p is the pressure (in bar),
- A is the cross-sectional area of the tubular reactor (in $\mathrm{m^2}$),
- U is the overall heat transfer coefficient (in $\mathrm{J/(m^2\ s\ K)}$).

The equations in 16 can be simplified by writing the vector f in terms of an orthonormal basis with $\|F\| = \sqrt{7} u_7$. Define

$$U := \begin{pmatrix} u_1 \\ \vdots \\ u_7 \end{pmatrix} \tag{20}$$

and C_v^u and C_p^u to be such that

$$C_v^u \begin{pmatrix} u_1 \\ \vdots \\ u_7 \end{pmatrix} = C_v F \text{ and } C_p^u \begin{pmatrix} u_1 \\ \vdots \\ u_7 \end{pmatrix} = C_p F. \tag{21}$$

Then, it can be shown that

[1] In the sequel, x and y in Sections 3 and 4 are refered to as t and z without loss of generality.

$$Me_i = \begin{cases} e_i, & i = 1, \ldots, 6 \\ -\dfrac{u_1}{u_7}e_1 - \cdots - \dfrac{u_6}{u_7}e_6, & i = 7 \\ -\dfrac{u_1}{T}e_1 - \cdots - \dfrac{u_7}{T}e_7 + \dfrac{C_v^u U}{\sqrt{7}RTu_7}e_8, & i = 8 \end{cases} \tag{22}$$

and

$$Ne_i = \begin{cases} \dfrac{RT}{Ap}e_i, & i = 1, \ldots, 6 \\ \dfrac{RT}{Ap}e_7 + \dfrac{\sqrt{7}RT}{Ap}e_8, & i = 7 \\ \dfrac{C_p^u U}{Ap}e_8, & i = 8 \end{cases} \tag{23}$$

Hence, the left-hand side of (16) for

$$u := \begin{pmatrix} u_1 \\ \vdots \\ u_7 \\ T \end{pmatrix} \tag{24}$$

can be written as

$$Au_t + Bu_x, \tag{25}$$

where

$$A = \begin{pmatrix} 1 & & -\dfrac{u_1}{u_7} & -\dfrac{u_1}{T} \\ & \ddots & \vdots & \vdots \\ & & 1 & -\dfrac{u_6}{u_7} & -\dfrac{u_6}{T} \\ & & & 0 & -\dfrac{u_7}{T} \\ & & & 0 & \dfrac{C_v^u U}{\sqrt{7}RTu_7} \end{pmatrix} \tag{26}$$

and

$$B = \begin{pmatrix} \dfrac{RT}{Ap} & & & \\ & \ddots & & \\ & & \dfrac{RT}{Ap} & \\ & & \dfrac{\sqrt{7}RT}{Ap} & \dfrac{C_p^u U}{Ap} \end{pmatrix}, \tag{27}$$

where the blank entries refer to zeroes.

This coordinate transformation results in a system of equations which has the same form as in (2) but has a much simplier matrix structure for the determination of the generalized eigenvalues and left-generalized eigenvectors in (12).

In particular, the analytical solution to $\det(A - \sigma B) = 0$ is

$$\sigma_k = \frac{Ap}{RT}, \qquad k = 1, \cdots, 6, \tag{28}$$

$$\boldsymbol{l}_1 = \begin{pmatrix} 1 \\ 0 \\ \vdots \\ 0 \end{pmatrix}, \ \ldots, \ \boldsymbol{l}_6 = \begin{pmatrix} 0 \\ \vdots \\ 0 \\ 1 \\ 0 \\ 0 \end{pmatrix} \tag{29}$$

and σ_7 and σ_8 solve

$$\det \begin{pmatrix} -\sigma\dfrac{RT}{Ap} & -\dfrac{u_7}{T} \\ -\sigma\dfrac{\sqrt{7}RT}{Ap} & \dfrac{C_v^u U}{\sqrt{7}RTu_7} - \sigma\dfrac{C_p^u U}{Ap} \end{pmatrix} = 0, \tag{30}$$

which can be written as

$$\frac{RTC_p^u U}{(Ap)^2}\sigma^2 - \left(\frac{C_v^u U}{\sqrt{7}Apu_7} - \frac{\sqrt{7}Ru_7}{Ap}\right)\sigma = 0, \tag{31}$$

whose solutions are

$$\sigma_7 = 0, \qquad \sigma_8 = \frac{Ap}{RTC_p^u U}\left(\frac{C_v^u U}{\sqrt{7}u_7} - \sqrt{7}Ru_7\right). \tag{32}$$

Since the eigenvalues are known, computing the left-generalized eigenvectors $\boldsymbol{l}_7$ and $\boldsymbol{l}_8$ is almost straightforward.

The above expressions mean that the floating point operations and memory locations for determining the generalized eigenvalues and left-generalized eigenvectors for the ethanol steam reforming model are much lower than the $\mathcal{O}(n^3)$ and $\mathcal{O}(n^2)$ estimates given in Section 4 for general first-order quasi-linear vector hyperbolic equations. In particular, both floating point and memory costs are negligible.

The online computational cost of solving the resulting system of ODEs (15) directly or incorporating them as constraints into a nonlinear model predictive control algorithm is also much lower for the ethanol steam reforming model than for the general equations. The σ_k and $\boldsymbol{l}_k$ in (15), as well as the matrix A and vector g, are simpler known explicit functions of the x, y, and u at any point in a dynamic simulation, so the computational cost is of incorporating the $2m$ ODEs in (15) into a nonlinear model predictive control algorithm would be very low (see, for example, Shang et al. (2004) and Munusamy et al. (2013)), e.g., by using a first-order forward Euler-type numerical scheme. The mathematical structure of the equations and the clear separation of time scales of the generalized eigenvalues suggest that further computational efficiency would be obtained by using a splitting scheme, e.g., by solving the ODEs for

$$\begin{pmatrix} u_7 \\ T \end{pmatrix} \tag{33}$$

and

$$\begin{pmatrix} u_1 \\ \vdots \\ u_6 \end{pmatrix} \tag{34}$$

alternatively.

The characteristic equations derived above have a quite different mathematical structure than what was reported in Reyero et al. (2020). The theoretical analysis presented here is much more complicated, but removes two approximations used in the past analysis that reduced the nonlinearity compared to the original model equations for the nonlinear distributed parameter system. As the mathematical reformation herein does not introduce any model error, the full nonlinearity of the original singular distributed parameter systems (2) is captured and can be exploited when incorporated into a nonlinear model predictive control algorithm.

6. CONCLUSIONS

This article derives a reduced-order model for an ethanol steam reformer that introduces no approximations in converting a system of singular nonlinear distributed parameter systems to a system of nonlinear ordinary differential equations for computing the states needed in nonlinear model predictive control formulations. The low online computational cost of the model equations is sufficiently low (only $2m$ ordinary differential equations) to enable a mechanistic model to be employed in real-time control calculations while taking input, state, and output constraints explicitly into account.

Although the analysis in this article was motivated by the ethanol steam reforming, a similar spatio-temporal analysis and control-relevant reduced-order model construction is expected to be extendable to other tubular reactors, membrane systems, and chromatography systems in which the effects of advection on the spatio-temporal dynamics are much stronger than the effects of diffusive phenomena. Such dynamical systems with high Peĉlet number commonly arise in chemical process control applications. The overall approach should also be extendable to nonlinear distributed parameter systems with additional coordinates including intrinsic properties such as particle number, mass, or age.

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