CFD Modeling and Control of a Steam Methane Reforming Reactor

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Abstract

This work initially focuses on developing a computational fluid dynamics (CFD) model of an industrial-scale steam methane reforming reactor (reforming tube) used to produce hydrogen. Subsequently, we design and evaluate three different feedback control schemes to drive the area-weighted average hydrogen mole fraction measured at the reforming tube outlet ($\bar{x}_{H_2}^{outlet}$) to a desired set-point value ($\bar{x}_{H_2}^{set}$) under the influence of a tube-side feed disturbance. Specifically, a CFD model of an industrial-scale reforming tube is developed in ANSYS Fluent with realistic geometry characteristics to simulate the transport and chemical reaction phenomena with approximate representation of the catalyst packing. Then, to realize the real-time regulation of the hydrogen production, the manipulated input and controlled output are chosen to be the outer reforming tube wall temperature profile and $\bar{x}_{H_2}^{outlet}$ respectively. On the problem of feedback control, a pro-

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portional (P) control scheme, a proportional-integral (PI) control scheme and a control scheme combining dynamic optimization and integral feedback control to generate the outer reforming tube wall temperature profile based on $\bar{x}_{H_2}^{set}$ are designed and integrated into real-time CFD simulation of the reforming tube to track $\bar{x}_{H_2}^{set}$. The CFD simulation results demonstrated that feedback control schemes can drive the value of $\bar{x}_{H_2}^{outlet}$ toward $\bar{x}_{H_2}^{set}$ in the presence of a tube-side feed disturbance, and can significantly improve the process dynamics compared to the dynamics under open-loop control.

Key words: Steam methane reforming, CFD modeling, process dynamics, process identification, process control, distributed parameter systems

1. Introduction

The steam methane reforming (SMR) process, which produces bulk hydrogen gas from methane through catalytic reactions, is the most common commercial method for industrial hydrogen production. A general industrial-scale SMR process can be described by the schematic of Fig. 1. The steam methane reformer (for simplicity, it is denoted as “reformer” in the following text) is the core unit in a SMR process which has a process (tube) side and a furnace side that interact through heat exchange through the walls of reforming reactors (for simplicity, they are denoted as the “reforming tubes”). In the furnace side, combustion of the furnace-side feed, usually a mixture of methane, hydrogen, carbon dioxide, carbon monoxide and air, heats the reforming tubes via radiative heat exchange; inside the reforming tubes, catalytic reactions take place, converting steam and methane into hydrogen and carbon oxides (including CO and CO_2). A traditional

14 top-fired, co-current furnace usually includes top burners which are fed with the
 15 furnace-side feed, refractory walls enveloping the combustion products, flue gas
 16 tunnels transporting the flue gas out of the reformer, and reforming tubes.

Figure 1: Steam methane reforming process diagram Othmer.

17 For the last 50 years, extensive work has been performed on the development
 18 of first principles reformer models. The mathematical modeling methodology of
 19 the complete reformer was first proposed and developed in the 1960s McGreavy and Newmann
 20 (1969). With the improved understanding of both physical and chemical phenom-
 21 ena inside the reformer, more comprehensive mathematical models have been de-
 22 veloped considering more detailed and precise radiation mechanisms, combus-

tion models, flue gas flow patterns, SMR reaction kinetics and packed bed reactor models Latham (2008). However, solving these complete reformer models is computationally expensive due to the increasing complexity of the fundamental nonlinear partial differential equations describing reformer physico-chemical phenomena. Also, for large reformers with complicated geometry, the geometry characteristics and various boundary conditions Latham (2008) make mathematical modeling very difficult.

On the other hand, with the dramatic increase of computing power, computational fluid dynamics (CFD) modeling has become an increasingly important platform for reformer modeling and design, combining physical and chemical models with detailed representation of the reformer geometry. When compared with first-principles modeling, CFD is a modeling technique with powerful visualization capabilities to deal with various geometry characteristics and boundary conditions. Moreover, CFD modeling provides flexibility to modify design parameters without the expense of hardware changes which brings large economic and time savings Uriz et al. (2013). CFD technology has been successful in carrying out the simulation of industrial furnaces Baburić et al. (2005); Han et al. (2006); Stefanidis et al. (2006); Noor et al. (2013) and SMR tube reactors, i.e., reforming tubes modeled as packed-bed reactors Calis et al. (2001); Behnam et al. (2012); Dixon (2014); Guardo et al. (2004). Specifically, recent attempts to use CFD modeling to characterize the physico-chemical phenomena of transport and reaction processes inside reforming tubes have been done exclusively on a microscopic or bench-scale level, e.g., the effect of catalyst orientation on catalytic

performance is investigated with a CFD model of a single catalyst particle Dixon (2014), and the validation of CFD simulation results to experimental data is performed with a CFD model of a bench-scale reforming tube Behnam et al. (2012). In the present work, we focus on an industrial-scale reforming tube, i.e., the external diameter, internal diameter and exposed length of the reforming tube are 14.6 *cm*, 12.6 *cm* and 12.5 *m* respectively. It is noteworthy that the CFD model of the industrial-scale reforming tube in this work is developed based on a unit currently employed in a commercial plant. Therefore, the CFD model has the geometry characteristics of an industrial reforming tube, i.e., the length and inner and outer tube radii, includes an industrially relevant representation of the catalyst network, and incorporates appropriate boundary conditions including the reforming tube outer wall temperature and tube-side feed conditions. A major purpose of the CFD modeling in this work, in addition to its purpose as a means for evaluating various control strategies, is to propose a method of industrial reforming tube modeling with considerable modeling accuracy that can help evaluate various modeling assumptions usually employed.

The production rate of hydrogen fuel from a typical SMR process strongly depends on the operating temperature of the furnace, where the aforementioned reforming tubes are encapsulated, and more specifically the outer reforming tube wall temperature. Because of the endothermic nature of SMR reactions, a higher outer reforming tube wall temperature theoretically results in a higher production rate of hydrogen fuel. On the other hand, operating the reforming tubes at excessively high temperature can lead to disastrous consequences and significant

capital loss. Particularly, the formation of carbon on the catalyst surface and on the inner reforming tube surface prevents the reactants from entering the catalyst active sites and reduces the rate of heat transfer to the tube-side gas mixture respectively. As a result, the reaction progress and hydrogen production might be disrupted. Additionally, the expected lifetime of reforming tubes is extremely sensitive to changes in operating temperatures, i.e., an increase in tube wall temperature of 20 K can reduce the tube lifetime by half Pantoleontos et al. (2012) Latham (2008). Moreover, reforming tubes are one of the most expensive plant components as they account for approximately 10% of the capital cost of an entire hydrogen plant Latham (2008), and the total replacement of a typical industrial-scale reformer is expected to cost 5 – 8 million USD Pantoleontos et al. (2012). Consequently, the outer reforming tube wall temperature is required to be closely monitored and kept slightly below the design tube wall temperature in industrial standard operation in order to eliminate the potential stress-to-rupture of tube materials Beyer et al. (2005). Through CFD simulation, these above critical issues can be detected and predicted, and corresponding changes can be applied to improve the reforming tube design and operating parameters.

Motivated by the above considerations, we initially develop a CFD model of an industrial-scale reforming tube in ANSYS Fluent with realistic tube geometry characteristics to simulate the transport and reaction phenomena with an approximate representation of the catalyst packing inside the reforming tube. Next, we utilize publicly available SMR plant data to construct the proper boundary conditions for the reforming tube inlet, the reforming tube outlet and the outer reform-

ing tube wall, so that the simulation results and the available industrial plant data
 are consistent Latham (2008). Due to the high Reynolds number of the tube-side
 gas mixture, significant pressure gradients across the reforming tube and well-
 acknowledged radial gradients near the reforming tube inlet, the standard $k - \epsilon$
 turbulence model with enhanced wall treatment is implemented. Furthermore, an
 intrinsic SMR reaction kinetic model Xu and Froment (1989), widely accepted in
 academia and industry, is used to derive the chemical conversion rate equations,
 which account for the effect of internal and external diffusion limitations on the
 observed rates as well as for the presence of the catalyst particles inside the re-
 forming tube. Subsequently, to adjust the hydrogen production of the reforming
 tube in real-time, we propose the design and implementation of feedback control
 schemes into the CFD model. Specifically, the manipulated input and controlled
 output are chosen to be the outer reforming tube wall temperature and the area-
 weighted average hydrogen mole fraction measured at the reforming tube outlet
 ($\bar{x}_{H_2}^{outlet}$) respectively, and the objective is to drive $\bar{x}_{H_2}^{outlet}$ to the desired set-point
 ($\bar{x}_{H_2}^{set}$) under the influence of a tube-side feed disturbance. On the problem of
 feedback control, firstly, a proportional (P) control scheme and a proportional-
 integral (PI) control scheme generating the outer reforming tube wall temperature
 trajectory based on the desired $\bar{x}_{H_2}^{set}$ are designed to realize the closed-loop CFD
 simulation. Finally, motivated by industrial concerns, we also design a feedback
 control scheme combining dynamic optimization and integral feedback control.
 The control performance of these three control schemes is evaluated with respect
 to tracking the set-point, improving the speed of the closed-loop responses and

115 compensating for the effect of the tube-side feed disturbance on $\bar{x}_{H_2}^{outlet}$.

116 **2. Single Reforming Tube Model**

117 *2.1. Industrial Level Geometry*

118 The reforming tube investigated in this work belongs to an industrial-scale
119 top-fired, co-current reformer designed by Sela Fluid Processing Corporation
120 (Figs. 2-3). The furnace of this reformer contains seven rows of forty-eight re-
121 forming tubes whose external diameter, internal diameter and exposed length
122 are 14.6 cm, 12.6 cm and 12.5 m respectively. Each reforming tube is tightly
123 packed with specially designed nickel oxide over alpha alumina support (i.e.,
124 $Ni/\alpha-Al_2O_3$) catalyst particles, which not only facilitate the formation of hydro-
125 gen fuel from steam and methane through the highly endothermic SMR reactions,
126 but also play a role as an intermediate medium to enhance the rate of heat transfer
127 to the tube-side gas mixture. These rows of tubes are separated by eight rows of
128 twelve burners (Fig. 2) which are fed with the furnace-side gas composed of three
129 parts, i.e., natural gas, combustion air (Ar , N_2 and O_2) and tail gas (CO and H_2).
130 Based on the composition of a typical furnace-side gas that is given in Table 1, the
131 combustion is a fuel lean process so that the fuel can be completely combusted
132 over a flame length of 4.5 – 6 m releasing the thermal energy needed to drive the
133 highly endothermic SMR reactions Latham (2008). The thermal energy released
134 by the combustion of furnace fuel is transferred to the reforming tubes predomi-
135 nantly by radiation inside the high-temperature furnace chamber. At the bottom of
136 the furnace, the rows of tubes are separated by the rectangular intrusions known
137 as flue gas tunnels or coffin boxes (Fig. 3). The flue gas tunnels extend from the

front to the back of the furnace with a height of 2.86 *m* from the floor and allow the furnace flue gas to exit the furnace. Thirty-five extraction ports are distributed in a row along the sides of each flue gas tunnel. The furnace flue gas enters the tunnels from the furnace chamber through the extraction ports and then exits the furnace through the front openings of the coffin boxes. It is worth noting that the dynamics of all aforementioned components are tightly coupled inside the furnace during operation.

Under ideal operating conditions, the environments surrounding all reforming tubes of the SMR furnace unit are very similar, such that it is necessary to model only a single reforming tube to characterize the conditions in all 336 reforming tubes. Thus, the scope of the modeling part in this work only aims to develop a CFD model with realistic dimensions, geometry and characteristics of one industrial-scale steam methane reforming tube, and in this regard, we develop and evaluate all essential simulation settings of the reforming tube model to accomplish the modeling objective in the following sections. By focusing on one reforming tube, we are able to accomplish the primary objective of this work, which is presented later, and which is to demonstrate that real-time feedback control schemes can be incorporated into the CFD model of an industrial-scale reforming tube to build closed-loop systems that have desired dynamic response behavior and produce a desired amount of hydrogen.

2.2. *Tube Geometry and Meshing*

Mesh quality is the most critical issue for accurate and successful CFD modeling, i.e., a low quality mesh requires the most robust CFD solver and significantly

Table 1: Furnace-side inlet operating conditions.

Pressure (kPa)	132.4
Temperature (K)	524
Flow rate (kg/s)	1.08
x_{CH_4}	0.0517
x_{H_2O}	0.0306
x_{CO}	0.0211
x_{H_2}	0.0540
x_{O_2}	0.1530
x_{Ar}	0.0077
x_{N_2}	0.5793

161 greater computing resources to determine a converged solution. Due to the ax-
 162 isymmetric geometry property of the reforming tube as shown in Fig. 4(a), a two-
 163 dimensional ($2D$) axisymmetric reforming tube geometry and its corresponding
 164 mesh structure were employed. The $2D$ axisymmetric reforming tube mesh which
 165 is shown in Fig. 4(b) was constructed in the meshing software ICEM-CFD. The
 166 $2D$ axisymmetric reforming tube mesh only contains 24690 quadrilateral cells,
 167 with a 100% orthogonal quality. Also, the CFD simulation of the single reform-
 168 ing tube only performs calculations for half of the tube mesh considering its $2D$
 169 axisymmetric properties. Consequently, this $2D$ axisymmetric reforming tube
 170 mesh brings large computational efficiency over a three-dimensional reforming
 171 tube CFD model.

172 In terms of mesh size of this single reforming tube, open-loop simulation
 173 results of CFD models with different mesh sizes indicate that 24690 cells are
 174 enough to represent this specific tube, and that the further increase of the mesh

Figure 2: The top view of an industrial-scale top-fired, co-current reformer with 336 reforming tubes, for which each reforming tube is denoted by a smaller circle, and 96 burners, for which each burner is denoted by a larger circle.

size achieves identical results but requires significantly higher computation time.

For this reforming tube CFD model, boundary layer design is very critical to the modeling of the heat convection from the inner reforming tube wall to the tube-side gas mixture and the heat conduction from the inner reforming tube wall to the catalyst particles. To calculate the first node height from the inner reforming tube wall for the boundary layer design, NASA's Viscous Grid Spacing Calculator Aeronautics and Administration is adopted based on a suitable Y^+ value Inc. (Nov. 2013a). For this specific single reforming tube geometry, five nodes are ap-

Figure 3: The front view of an industrial-scale top-fired, co-current reformer. It is important to note that the refractory wall is modeled to be transparent, so that the interior components can be seen. Specifically, eight rectangular boxes located on the floor represent the flue gas tunnels, eight frustums of cones located on the ceiling represent the corresponding rows of burners and seven slender rectangles connecting the ceiling and floor represent the corresponding rows of reforming tubes.

183 plied in the boundary layer at the inner reforming tube wall as requested by the
 184 two-equation $k - \epsilon$ turbulence model with the enhanced wall treatment function
 185 (which will be discussed below). The first node height is $8.26 \times 10^{-4} \text{ m}$, and the
 186 node spacing ratio in the boundary layer is 6 : 5. The detailed mesh structure for

187 the boundary layer design is demonstrated in Fig. 4(b). We note here that NASA's
188 Viscous Grid Spacing Calculator Aeronautics and Administration uses fixed vis-
189 cosity, pressure and temperature values for the fluid properties which may not be
190 suitable for most SMR calculations. Through a decompiling of the calculator, a
191 revised algorithm based on the original calculator is developed for our specific
192 inlet conditions of the tube-side gas mixture.

Figure 4: Two-dimensional axisymmetric reforming tube geometry (Fig. 4(a)) and mesh structure (Fig. 4(b)).

193 2.3. *Kinetic Model*

194 The conversion rates of reactants, i.e., steam and methane, into products, i.e.,
195 hydrogen fuel, and the direction of the reforming reactions and the water-gas shift
196 reaction under different conditions (reactant concentration, temperature and pres-
197 sure) must be accurately accounted for by using a reaction kinetics model. In
198 a reforming tube, the reforming reactions and water-gas shift reaction occur at
199 the catalyst active sites. In particular, reactants need to diffuse from the bulk
200 tube-side gas mixture to the surface of the catalyst particle and then into the cat-
201 alyst pores; after the formation of the products, products need to desorb from the
202 catalyst cores and reenter the bulk tube-side gas mixture. However, a reaction
203 kinetics model that provides a detailed treatment of these catalyst-specific phe-
204 nomena would be unnecessarily complex. Additionally, the catalyst particles and
205 the detailed packing pattern inside an industrial-scale packed-bed reactor are not
206 explicitly modeled in this study.

207 To deal with this issue, in this single reforming tube CFD model, an intrinsic
208 SMR reaction model Xu and Froment (1989) is used to describe the reactions
209 happening inside the reforming tube. The SMR reaction kinetics are given in
210 Eq. 1 below which is widely adopted in both CFD modeling and mathematical
211 modeling of reforming tubes:

$$CH_4(g) + H_2O(g) \rightleftharpoons CO(g) + 3H_2(g),$$

$$r_1 = \frac{k_1}{p_{H_2}^{2.5}} \left(p_{CH_4} p_{H_2O} - \frac{p_{H_2}^3 p_{CO}}{K_1} \right) / DEN^2 \quad (1a)$$

$$CO(g) + H_2O(g) \rightleftharpoons CO_2(g) + H_2(g),$$

$$r_2 = \frac{k_2}{p_{H_2}} \left(p_{CO} p_{H_2O} - \frac{p_{H_2} p_{CO_2}}{K_2} \right) / DEN^2 \quad (1b)$$

$$CH_4(g) + 2H_2O(g) \rightleftharpoons CO_2(g) + 4H_2(g),$$

$$r_3 = \frac{k_3}{p_{H_2}^{3.5}} \left(p_{CH_4} p_{H_2O}^2 - \frac{p_{H_2}^4 p_{CO_2}}{K_3} \right) / DEN^2 \quad (1c)$$

$$DEN = 1 + \frac{K_{H_2O} p_{H_2O}}{p_{H_2}} \quad (1d)$$

$$+ K_{CO} p_{CO} + K_{H_2} p_{H_2} + K_{CH_4} p_{CH_4}$$

212 where p_{H_2} , p_{CH_4} , p_{H_2O} , p_{CO} and p_{CO_2} are the partial pressures of H_2 , CH_4 , H_2O ,
 213 CO and CO_2 respectively in the bulk tube-side gas mixture inside the reforming
 214 tube, K_{H_2} , K_{CH_4} and K_{CO} are adsorption constants for H_2 , CH_4 and CO , re-
 215 spectively, K_{H_2O} is a dissociative adsorption constant of H_2O , k_1 , k_2 and k_3 are
 216 rate coefficients of the SMR reactions and DEN is a dimensionless parameter. To
 217 realize this reaction kinetic model in CFD simulations by Fluent, each reversible
 218 reaction in Eq. 1 is split into two irreversible reactions. The reaction rates in Eq. 1
 219 are implemented in Fluent by designing a user defined function (UDF) file. Since
 220 these intrinsic reaction kinetics do not consider the internal and external diffusion
 221 resistances of the catalyst particles, the reaction rates in Eq. 1 are multiplied by
 222 an effectiveness factor, 0.1 Wesenberg and Svendsen (2007), to account for the
 223 overall diffusion effects on the intrinsic reaction rates.

224 2.4. Compressible Gas Flow

225 The tube-side inlet operating conditions of the reforming tubes are given in
 226 Table 2 Latham (2008). Based on the inlet conditions of the tube-side gas mixture,
 227 the Mach number is determined to be greater than 0.3, and therefore, the density
 228 variations of the fluid flow due to high static pressure cannot be ignored. From
 229 the point of view of CFD simulation, when a pressure-based solver is used, like
 230 the one we chose for the simulation in this work, an accurate gas state equation is
 231 very critical to simulation accuracy. Based on this consideration, the compressible
 232 ideal gas state equation is adopted to describe the compressibility of the tube-side
 233 gas mixture.

Table 2: Process-side gas inlet operating conditions.

Pressure (kPa)	3038.5
Temperature (K)	887
Flow rate (kg/s)	0.1161
x_{CH_4}	0.2487
x_{H_2O}	0.7377
x_{CO}	0.0001
x_{H_2}	0.0018
x_{CO_2}	0.0117

234 A pressure-based solver that enables the pressure-based Navier-Stokes so-
 235 lution algorithm Inc. (Nov. 2013b) is chosen over a density-based solver. The
 236 pressure-based solver is more suited for a wider range of physical models and
 237 has features which are unavailable to the density-based solver, e.g., the physical
 238 velocity formulation for porous media which is adopted by this work to simulate

the flow through the catalyst network in the reforming tube (this will be discussed later). It also provides more freedom for the simulations while converging to the same results obtained by the density-based solver.

2.5. Porous Zone Design

Table 3: Johnson Matthey’s Katalco 23 – 4Q catalyst properties.

Density, ρ_c	3960 kg/m^3
Heat Capacity, $C_{p,c}$	880 $J/(kg \cdot K)$
Thermal Conductivity, k_c	33 $W/(m \cdot K)$
Particle Diameter, D_p	3.5 mm (average)

Pressure drop is significant in industrial-scale reforming tubes when the tube-side gas mixture flows through the catalyst network made of many tightly packed catalyst particles. To estimate the pressure drop across the porous zone in the CFD modeling of turbulent flows, a semi-empirical expression, the Ergun equation Ergun and Orning (1949), which is applicable over a wide range of Reynolds numbers and for many packing patterns, is adopted as follows:

$$\frac{\Delta P}{L} = \frac{150\mu}{D_p^2} \frac{(1-\gamma)^2}{\gamma^3} v_\infty + \frac{1.75\rho}{D_p} \frac{(1-\gamma)}{\gamma^3} v_\infty^2 \quad (2)$$

where ΔP is the pressure drop through the porous media, L is the depth of the porous media, μ is the viscosity of the fluid, γ is the porosity of the packed bed, v_∞ is the bulk velocity of the fluid, ρ is the density of the porous media, $\frac{150\mu}{D_p^2} \frac{(1-\gamma)^2}{\gamma^3}$ is the viscous resistance coefficient and $\frac{1.75\rho}{D_p} \frac{(1-\gamma)}{\gamma^3}$ is the inertial resistance coefficient. We assume that the viscous and inertial resistance coefficients are defined along the direction vectors $v_1 = [1, 0]$ (i.e., the principal axis direction)

255 and $v_2 = [0, 1]$ (i.e., the radius direction) in the Cartesian two-dimensional (2D)
256 coordinate system.

257 The modeling objective of this work is to develop a CFD model of an industrial-
258 scale reforming tube for a commercial plant. Thus, a common, readily available
259 commercial catalyst, i.e., Johnson Matthey's Katalco 23–4Q Matthey also known
260 as $Ni/\alpha - Al_2O_3$, is chosen for this purpose, and its properties are shown in Ta-
261 ble 3. It is noted that the intrinsic kinetic model used Xu and Froment (1989)
262 was developed for a $Ni/MgAl_2O_4$ catalyst, i.e., the catalyst support material is
263 different than that utilized in this work. We considered the potential effects of
264 the support material on the catalytic activity of the catalyst prior to selecting the
265 kinetic model developed in Xu and Froment (1989) based on the work conducted
266 by Jeong et al. (2006). First of all, the reduction peaks of both the $Ni/MgAl_2O_4$
267 and $Ni/\alpha - Al_2O_3$ catalyst types are in the neighborhood of $\sim 1083\text{ K}$, which
268 means that they require similar heat input to become activated. Even though
269 $Ni/MgAl_2O_4$ is more active at lower temperatures than $Ni/\alpha - Al_2O_3$, we specu-
270 late that both catalyst types have similar catalytic activity in our operating temper-
271 ature range. Additionally, although the estimated surface area of $Ni/MgAl_2O_4$ is
272 higher than that of $Ni/\alpha - Al_2O_3$ per unit mass of the catalyst, due to the large
273 porosity of the packed bed inside the reforming tube, i.e., the void volume is ap-
274 proximately 60.9% of the reactor volume, and small inner reforming tube diameter
275 of 0.126 m, the mass of catalyst inside a single reforming tube is not significant.
276 As a result, we decided to adopt the kinetic model developed in Xu and Froment
277 (1989) in the CFD model to describe the chemistry inside the reforming tube us-

ing the Johnson Matthey’s Katalco 23 – 4Q catalyst. Finally, the consistency of our simulation results in later sections with the data with which it is validated also demonstrates the effectiveness of the choice of the $Ni/\alpha - Al_2O_3$ catalyst for the kinetic model based on a $Ni/MgAl_2O_4$ catalyst. Additionally, from Table 2 and Table 3, the mass of the catalysts per reforming tube is 242 kg, and the space-time of the tube is about 2080 seconds.

2.6. Reforming Tube Wall Boundary Conditions

For an industrial-scale reformer, the outer reforming tube wall temperature is usually in the range of 1100 K to 1178 K Latham (2008). In this single reforming tube simulation, to construct the temperature boundary condition of the outer reforming tube wall, the available plant data Latham (2008) are fit with a fourth order polynomial function by using a least squares linear regression method. The result of the fit shown in Fig. 5 is

$$T_{Wall}(x) = [x^4 \ x^3 \ x^2 \ x^1 \ x^0] \begin{bmatrix} -0.0221 \\ 0.8003 \\ -10.734 \\ 64.416 \\ 997.16 \end{bmatrix} \quad (3)$$

where $T_{Wall}(x)$ is the outer reforming tube wall temperature at a location x (m) away from the reforming tube inlet, $x = 0$ m. Using the boundary condition in Eq. 3, the reforming tube here is modeled as a heat sink, and thermal energy is transferred from the outer reforming tube wall to the inner reforming tube wall by heat conduction. The corresponding tube material properties are listed in Table 4 Davis (2000) which assumes that all the tube properties are temperature

297 independent and the values at $T = 1144 \text{ K}$ are adopted in the CFD simulation.
 298 However, a constant outer reforming tube wall temperature profile is neither suf-
 299 ficient to maintain the current $\bar{x}_{H_2}^{outlet}$ when the reforming tube operating condi-
 300 tion is subjected to a change in the tube-side mass flow rate (e.g., tube-side feed
 301 disturbance), nor capable of driving the current $\bar{x}_{H_2}^{outlet}$ to a new desired $\bar{x}_{H_2}^{set}$. To
 302 fulfill the overall scope of this work, which is to design and implement feedback
 303 control schemes to drive $\bar{x}_{H_2}^{outlet}$ to the desired $\bar{x}_{H_2}^{set}$ under the influence of a tube-
 304 side feed disturbance, additional temperature profiles are constructed based on
 305 the most essential criterion that the shapes (in the axial direction) of these addi-
 306 tional temperature profiles have to match that of the original outer reforming tube
 307 wall temperature profile (owing to the overall shape and location of burner flames
 308 which does not change appreciably with time). The additional profiles are given
 309 by the following fourth-order polynomial,

$$T_{Wall}(x) = [x^4 \ x^3 \ x^2 \ x^1 \ x^0] \begin{bmatrix} -0.0221 \\ 0.8003 \\ -10.734 \\ 64.416 \\ T_{Wall}^{max} - 151.83 \end{bmatrix} \quad (4)$$

310 where T_{Wall}^{max} is the maximum outer reforming tube wall temperature of the tem-
 311 perature profile. Eq. 4 allows one to construct an entire outer reforming tube wall
 312 temperature profile along the reforming tube length based on its maximum tem-
 313 perature value and consequently affect the $\bar{x}_{H_2}^{outlet}$.

314 2.7. Governing equations

315 In the microscopic view of the reforming tube, the physico-chemical phenom-
 316 ena of transport and reaction processes that generate hydrogen fuel from steam

Figure 5: Outer reforming tube wall temperature versus distance down the reforming tube for the CFD simulations, which depicts the direct comparison between the fourth order polynomial temperature profile, i.e., Eq. 3 and that of publicly available SMR plant data Latham (2008).

Table 4: Reforming tube material properties.

Density, ρ_t	7720 kg/m^3
Heat Capacity, $C_{p,t}$	$502 \text{ J/(kg} \cdot \text{K)}$ at 1144 K
Thermal Conductivity, k_t	$29.58 \text{ W/(m} \cdot \text{K)}$ at 1144 K
Emissivity, ϵ_t	0.85 at 1144 K

317 and methane are closely coupled and utterly complex. Initially, a convective mass
318 transfer process driven primarily by the reactant concentration gradients between
319 a flowing bulk gas mixture and an infinitesimally thin stationary layer around the
320 catalytic surface takes place. Next, a molecular diffusion process mainly driven by

Figure 6: Outer reforming tube wall temperature versus distance down the reforming tube for the CFD simulations, which represents the calculated outer reforming tube wall temperature profiles generated by Eq. 4 and the fitted outer reforming tube wall temperature profile (solid line) based on the publicly available SMR plant data Latham (2008)

the reactant concentration and temperature gradients (Eq. 6g) between an infinitely thin stationary layer around the catalyst surface and a catalyst medium allows the reactants to diffuse into the catalyst pores and then finally arrive at the catalyst active sites, where the endothermic SMR reactions take place. Upon the formation of the products, they immediately desorb from the catalyst active sites, diffuse back to the catalyst surface and eventually reenter the flowing bulk gas mixture. It is worth noting that the series of consecutive molecular-level elementary steps of the endothermic SMR reactions is still largely unknown. As a result, a kinetic model that provides a detailed treatment of these catalyst-specific

330 phenomena would be unnecessarily complex from the point of view of CFD mod-
331 eling; therefore, the kinetic model used in this paper is derived based on a widely-
332 accepted intrinsic SMR reaction kinetic model Xu and Froment (1989) to lessen
333 the computational burden without sacrificing the accuracy of the simulation re-
334 sults (Sec. 2.3).

335 In the macroscopic view of the reforming tube, the significant axial pressure
336 gradient across the reforming tube length and radial gradients near the reform-
337 ing tube entrance are well-acknowledged. This, in turn, motivates us to develop
338 two-dimensional (2D) governing equations that are capable of accounting for the
339 presence of the catalyst network formed by the solidly packed catalyst particles. It
340 is important to thoroughly understand the contribution of the catalyst network to
341 the SMR reactions in order to develop an appropriate process model. Specifically,
342 the catalyst network not only facilitates the formation of hydrogen fuel from the
343 naturally stable and slowly-reacting materials, i.e., steam and methane, but also
344 enhances the rate of convective thermal energy transfer from the heated outer re-
345 forming tube wall to the flowing bulk gas mixture by expanding its contact area.

346 Additionally, due to the intrinsic resistance properties of the catalyst network,
347 i.e., the inertial resistance and viscous resistance previously mentioned in Sec. 2.5,
348 the catalyst network acts as a physical obstacle that interferes with the flow of
349 the tube-side gas mixture to generate turbulence. This obstacle enhances mixing
350 efficiency to radially homogenize the moving fluid. Furthermore, under the in-
351 fluence of flow resistances induced by the catalyst network, the residence time of
352 all species in the reforming tube is also increased, which allows the SMR reac-

353 tions Xu and Froment (1989) to reach equilibrium prior to the process gas exiting
 354 the reforming tube. Based on the aforementioned considerations, the 2D govern-
 355 ing equations, i.e., the continuity equation (Eq. 5) and the momentum (Eq. 6a),
 356 energy (Eq. 6b) and species material (Eq. 6c) balances, of the SMR process tak-
 357 ing place inside the reforming tube are formulated as follows, so that the presence
 358 of the catalyst network is explicitly accounted for:

$$\frac{\partial}{\partial t}(\gamma \rho_{fluid}) + \nabla \cdot (\gamma \rho_{fluid} \vec{v}) = 0 \quad (5)$$

$$\begin{aligned} \frac{\partial}{\partial t}(\gamma \rho_{fluid} \vec{v}) + \nabla \cdot (\gamma \rho_{fluid} \vec{v} \vec{v}) = & \quad (6a) \\ - \gamma \nabla P + \nabla \cdot (\gamma \vec{\tau}) + \gamma \vec{B}_f - \left(\frac{\gamma^2 \mu}{\alpha} \vec{v} + \frac{\gamma^3 C_2}{2} \rho |\vec{v}| \vec{v} \right) \end{aligned}$$

$$\frac{\partial}{\partial t}(\gamma \rho_{fluid} E_{fluid}) + \frac{\partial}{\partial t}((1 - \gamma) \rho_{solid} E_{solid}) \quad (6b)$$

$$\begin{aligned} & + \nabla \cdot (\vec{v}(\rho_{fluid} E_{fluid} + P)) = \\ & \nabla \cdot \left[k_{eff} \nabla T - \left(\sum_i h_i J_i \right) + \vec{\phi} \right] + \sum_i \Delta H_{rxn,i} \\ \frac{\partial}{\partial t}(\gamma \rho_{fluid} Y_i) + \nabla \cdot (\gamma \rho_{fluid} \vec{v} Y_i) = & - \nabla \cdot (\gamma \vec{J}_i) + R_i \quad (6c) \end{aligned}$$

with

$$\vec{v}_{superficial} = \gamma \vec{v} \quad (6d)$$

$$\vec{\tau} = \mu \left[(\nabla \vec{v} + \nabla \vec{v}^T) - \frac{2}{3} \nabla \cdot \vec{v} I \right] \quad (6e)$$

$$k_{eff} = \gamma k_{fluid} + (1 - \gamma) k_{solid} \quad (6f)$$

$$\vec{J}_i = \left(\rho_{fluid} D_{m,i} + \frac{\mu_t}{Sc_t} \right) \nabla Y_i - D_{T,i} \frac{\nabla T}{T} \quad (6g)$$

$$\begin{aligned} \vec{\phi} = 2\mu & \left[\left(\frac{\partial v_x}{\partial x} \right)^2 + \left(\frac{\partial v_y}{\partial y} \right)^2 - \frac{1}{3} (\nabla \cdot \vec{v})^2 \right] \\ & + \mu \left[\frac{\partial v_y}{\partial x} + \frac{\partial v_x}{\partial y} \right]^2 \quad (6h) \end{aligned}$$

359 where μ , μ_t , $\vec{v}$, $\vec{v}_{superficial}$, ρ_{fluid} and $\vec{B}_f$ are the molecular viscosity, turbulent vis-
360 cosity, physical velocity vector, superficial velocity vector, average density, and
361 body force of the bulk tube-side gas mixture respectively, γ , ρ_{solid} and α are the
362 porosity, density and absolute permeability of the catalyst network respectively,
363 $\frac{1}{\alpha}$ and C_2 represent the viscous resistance coefficient and inertial resistance coef-

364 ficient (previously introduced in Sec. 2.5), E_{fluid} and E_{solid} are the total energy
 365 of the tube-side gas mixture and of the catalyst network, P is the static pressure,
 366 $\sum_i \Delta H_{rxn,i}$ is the total thermal energy generated by the SMR reactions, k_{fluid} ,
 367 k_{solid} and k_{eff} are the thermal conductivities of the tube-side gas mixture, of the
 368 catalyst network and of the overall medium, Y_i , $D_{m,i}$, $D_{T,i}$, $\vec{J}_i$ and R_i are the mass
 369 fraction, mass diffusion coefficient, thermal diffusion coefficient, turbulent mass
 370 diffusion flux and overall rate of chemical reaction of species i , Sc_t is the turbulent
 371 Schmidt number and is set to 0.7 by default, and $\vec{\tau}$ and I are the stress tensor and
 372 unit tensor, respectively. It is important to note that the chemical rate equations are
 373 formulated to account for the effects of internal and external diffusion limitations
 374 on the observed rates as well as for the presence of the catalyst particles inside
 375 the reforming tube, and therefore, it would be unnecessary and incorrect to mul-
 376 tiply R_i in Eq. 6c and $\sum_i \Delta H_{rxn,i}$ in Eq. 6b by γ in an attempt to account for the
 377 effects of the catalyst network. Additionally, detailed microscopic transport and
 378 chemical reaction processes are described by the 2D governing equations, i.e.,
 379 $[\nabla \cdot (\sum_i h_i J_i)]$ in Eq. 6b represents the transport of enthalpy due to molecular
 380 diffusion, and $[(\nabla \vec{v} + \nabla \vec{v}^T) - \frac{2}{3} \nabla \cdot \vec{v} I]$ in Eq. 6e represents the effect of volume
 381 dilation.

382 Due to the high Reynolds number of the tube-side gas mixture, which is
 383 estimated to be ~ 5500 based on the inlet conditions of the tube-side feed as
 384 shown in Table 2, the semi-empirical standard two-equation turbulent kinetic
 385 energy and turbulent dissipation rate ($k - \epsilon$) model Jones and Launder (1972);
 386 Launder and Sharma (1974) developed from the Reynolds-averaged Navier-Stokes

387 (RANS) equations is employed to describe the complex turbulence phenomena in-
 388 side the reforming tube. The $k - \epsilon$ turbulence model presented in Eq. 7 below is
 389 applicable for a wide range of flows and is relatively computationally inexpensive,
 390 though it still yields reasonably accurate estimates, and it is relatively simple to
 391 implement and easy to converge from the point of view of CFD simulation. This
 392 model has the form:

$$\begin{aligned} \frac{\partial}{\partial t}(\gamma \rho_{fluid} k) + \nabla \cdot (\gamma \rho_{fluid} \vec{v} k) = \nabla \cdot \left[\gamma \left(\mu + \frac{\mu_t}{\sigma_k} \right) \nabla k \right] \\ + \gamma G_k + \gamma G_b - \gamma \rho_{fluid} \epsilon - \gamma Y_M \end{aligned} \quad (7a)$$

$$\begin{aligned} \frac{\partial}{\partial t}(\gamma \rho_{fluid} \epsilon) + \nabla \cdot (\gamma \rho_{fluid} \epsilon \vec{v}) = \nabla \cdot \left[\gamma \left(\mu + \frac{\mu_t}{\sigma_\epsilon} \right) \nabla \epsilon \right] \\ + \gamma C_{1\epsilon} \frac{\epsilon}{k} (G_k + C_{3\epsilon} G_b) - \gamma C_{2\epsilon} \rho \frac{\epsilon^2}{k} \end{aligned} \quad (7b)$$

$$\left\{ \begin{array}{l} \mu_t = \rho_{fluid} C_\mu \frac{k^2}{\epsilon} \\ C_{3\epsilon} = \tanh \left| \frac{v}{u} \right| \\ G_b = \beta g_i \frac{\mu_t}{Pr_t} \frac{\partial T}{\partial x_i} \\ G_k = -\rho u'_i u'_j \frac{\partial u_j}{\partial x_i} \\ \beta = -\frac{1}{\rho} \left(\frac{\partial \rho}{\partial T} \right)_P \end{array} \right.$$

393 where k and ϵ are the turbulent kinetic energy and turbulent dissipation rate re-
 394 spectively, G_k and G_b are the generation of turbulent kinetic energy due to the
 395 mean velocity gradients and buoyancy respectively, Y_M is the contribution of
 396 the fluctuating dilation in compressible turbulence to the overall dissipation rate,
 397 $\sigma_\epsilon = 1.0$ and $\sigma_k = 1.3$ are the turbulent Prandtl numbers for ϵ and k respectively,

398 $C_{1\epsilon} = 1.44$, $C_{2\epsilon} = 1.92$ and $C_\mu = 0.09$ are default constants of the standard
 399 $k - \epsilon$ model, $Pr_t = 0.85$ is the turbulent Prandtl number for energy, g_i is the
 400 component of the gravitational vector in the i th direction, β is the thermal ex-
 401 pansion coefficient, and v and u are the components of the flow velocity parallel
 402 and perpendicular, respectively, to the gravitational vector. It is worth noting that
 403 all default values of the aforementioned constants are determined empirically by
 404 experiments for fundamental turbulent flows, i.e., boundary layers and mixing
 405 layers, and have been verified to be suitable for a wide range of wall-bounded and
 406 free shear flow applications Inc. (Nov. 2013a). Furthermore, turbulent flows are
 407 significantly affected by the walls. The accuracy of the near-wall modeling de-
 408 cides the fidelity of numerical solutions. Specifically, it is in the near-wall region
 409 that the solution variables have large gradients, i.e., the momentum, material and
 410 energy fluxes have large magnitudes. Therefore, an accurate representation of the
 411 flow in the near-wall region is required for successful predictions of wall-bounded
 412 turbulent flows. For this single reforming tube CFD model, the enhanced wall
 413 treatment function in Fluent is applied as the near wall treatment method. The
 414 enhanced wall treatment ϵ -Equation is suitable for certain fluid dynamics espe-
 415 cially for those with lightly turbulent flow, and it only requires a few nodes in the
 416 boundary layer when using the $k - \epsilon$ model Inc. (Nov. 2013a).

417 2.8. Simulation Results

418 2.8.1. CFD Model of Single Reforming Tube: Results Comparison

419 Using a parallel computational environment with message passing interface
 420 technology, the simulation of the single reforming tube CFD model converges in

about 5 minutes with the steady solver in a 4-core CPU desktop computer. The steady-state results of the simulations are displayed in Figs. 7-8. We note here that due to the large length to diameter ratio of the reforming tube, in all plots of the simulation results, the radius is scaled up by 20 times which is convenient and is done for display purposes only. It is worth noting that the pressure profile in this simulation is radially uniform, which is the result of the direction definitions of the resistance coefficients in Section 2.5. In addition, based on the wall temperature profile in Fig. 5, the corresponding inner reforming tube wall temperature profile at the steady-state from the converged model is displayed in Fig. 9, and the corresponding steady-state heat flux profile through the tube wall is shown in Fig. 10.

Figure 7: Temperature profile from the reforming tube CFD simulation, where the outer reforming tube wall temperature profile (Fig. 5) is that fitted based on the available SMR plant data Latham (2008).

432 Reforming tube outlet data (not reported here for proprietary reasons) from
433 a typical hydrogen plant with the same tube-side inlet conditions, the same tube
434 geometric structure and similar catalyst and tube material properties as described
435 in Secs. 2.1, 2.4 and 2.5, and also the same wall temperature profile as that re-
436 ported in Fig. 5, is used to validate the CFD single tube model. The validation is
437 performed by comparing the data of the process gas at the reforming tube outlet
438 (Table 5) from the CFD simulation to that of the available proprietary industrial
439 plant. All mole fraction values in Table 5 are area-weighted average values at the
440 tube outlet. We found that our simulation results reported in Table 5 are very close

Figure 8: Hydrogen mole fraction profiles from the reforming tube CFD simulation, where the outer reforming tube wall temperature profile (Fig. 5) is that fitted based on the available SMR plant data Latham (2008).

441 to the plant data for temperature and species compositions.

442 Additionally, the average heat flux across the reforming tube wall produced by
 443 the CFD model is within the range of $45 - 90 \text{ kW/m}^2$ Rostrup-Nielsen (1984), a
 444 typical range reported in industry, and closely resembles that reported throughout
 445 literature. Namely, Dybkjaer reports the average value of 78.5 kW/m^2 Dybkjaer
 446 (1995) and Froment and Bischoff report the average value of 75.6 kW/m^2 Froment and Bischoff
 447 (1990). Moreover, the work of Pantoleontos *et al.* on modeling and optimiza-
 448 tion of a reforming reactor also restricts the average heat flux to be less than
 449 80.0 kW/m^2 Pantoleontos et al. (2012). Furthermore, the estimated average heat

flux of 65.6 kW/m^2 based on the outer and inner reforming tube wall temperature profiles, thermal conductivity of $106500 \frac{\text{J}}{\text{m}\cdot\text{h}\cdot\text{K}}$ and thickness of the reforming tube wall reported by Latham validates the accuracy of the CFD model Latham (2008).

Figure 9: Inner (dashed line) and outer (solid line) wall temperature profiles of the reforming tube.

Remark 1. The outlet tube-side mixture compositions are at their equilibrium state at the exit of the reactor which is determined by the outlet tube-side mixture temperature. Further, the average hydrogen mole fraction in the tube-side mixture measured at the tube outlet, $\bar{x}_{H_2}^{outlet}$, can be mathematically expressed as a function of the maximum reforming tube wall temperature, T_{Wall}^{max} . It is important to

Figure 10: Heat flux profile across the reforming tube wall.

459 note that T_{Wall}^{max} is not necessarily the temperature of the tube-side mixture at the
 460 outlet, which depends not only on T_{Wall}^{max} , but also depends on the thermal conduc-
 461 tivity of the reforming tube wall, the effective heat transfer coefficient between
 462 the tube-side mixture and the catalyst network, and especially, the rate of energy
 463 consumption by the endothermic SMR reactions. A major benefit of creating a
 464 CFD model of the industrial-scale reforming tube is that it allows relationships
 465 between important quantities within the reformer to be determined for control or
 466 other analysis purposes. For example, it allows the generation of the collection
 467 of data at constant tube-side feed conditions needed to construct the relationship
 468 between $\bar{x}_{H_2}^{outlet}$ and T_{Wall}^{max} used for control purposes later in this work. Addition-
 469 ally, the industrial-scale reforming tube CFD model creates more opportunities for

470 future investigations. Specifically, the CFD model allows us to access additional
471 information about the tube-side mixture at the reforming tube outlet, e.g., the area-
472 weighted average temperature of the tube-side mixture denoted by $\bar{T}_{Mixture}^{outlet}$, which
473 can be used to construct the relationship between $\bar{T}_{Mixture}^{outlet}$ and T_{Wall}^{max} to construct,
474 for example, a Gibbs reactor model.

475 Additional effort to validate the simulation result generated by the single re-
476 forming tube CFD model in order to declare with high certainty that the CFD
477 model can replace expensive traditional on-site parametric study, and to also demon-
478 strate that the tube-side gas mixture at the reforming tube outlet is at equilib-
479 rium conditions is conducted. Specifically, a Gibbs reactor model of a steady
480 state chemical and petroleum process simulation software known as *Pro/II* is
481 adopted to simulate the reforming tube to determine the species composition at
482 chemical equilibrium by solving the energy and material conservation equations
483 based on minimizing the Gibbs free energy of all chemical species involved in
484 this process. Furthermore, the greatest advantage of the Gibbs reactor model is
485 that it does not require the knowledge of chemical process in the reforming tube
486 including detailed reaction mechanism or kinetic model, which in turn validate
487 the global reaction mechanism and kinetic models of SMR process we imple-
488 mented in the single reforming tube CFD model. In this effort, the Gibbs reactor
489 model, which requires only two input parameters, i.e a Gibbs reactor feed stream,
490 which is identical to that of tube-side feed (shown in Table 2), and a Gibbs reactor
491 duty, which is set to the total thermal energy absorbed by the reforming tube of
492 341.045 kW, is simulated, and the corresponding simulation results are shown in

493 Table 5. The simulation results generated by the Gibbs reactor model are in close
 494 agreement with that generated by the single reforming tube CFD model, which
 495 in turn support the accuracy of the CFD model, validate our choice of the global
 496 reaction mechanism and kinetic model of SMR process, and also demonstrate that
 497 the tube-side gas mixture at the reforming tube outlet is at equilibrium conditions.

Table 5: Single reforming tube results.

	CFD Result	Pro/II	% Difference with respect to CFD result
ΔP (kPa)	212.83	N/A	N/A
P_{outlet} (kPa)	3044.0	3044.0	0.00
$\bar{x}_{H_2}^{outlet}$	0.4645	0.4650	0.11
$\bar{x}_{H_2O}^{outlet}$	0.3467	0.3452	0.43
$\bar{x}_{CH_4}^{outlet}$	0.0426	0.0422	0.94
$\bar{x}_{CO}^{outlet}$	0.0873	0.0869	0.46
$\bar{x}_{CO_2}^{outlet}$	0.0588	0.0607	3.23
Average heat flux ($\frac{kW}{m^2}$)	68.972	N/A	N/A
Total absorbed heat (kW)	341.045	341.055	~ 0.00

498 3. Feedback control design and closed-loop simulation results

499 3.1. Open-loop dynamics

500 We are primarily interested in the regulation of $\bar{x}_{H_2}^{outlet}$ when the reforming tube
 501 is subjected to tube-side feed disturbances, where the control objective is to main-
 502 tain the current $\bar{x}_{H_2}^{set}$, or when changes in industrial plant objectives occur, where
 503 the control objective is to attain a new $\bar{x}_{H_2}^{set}$.

504 Among all simulation settings described in Section 2, the outer reforming tube
 505 wall temperature profile is chosen as the single manipulated input due to the en-
 506 dothermic nature of the SMR reactions. As a result, $\bar{x}_{H_2}^{outlet}$ can be manipulated by
 507 adjusting the outer reforming tube wall temperature profile, which depends on the
 508 fuel flow rates to the burners around the reforming tube. It should be noted that
 509 the maximum value of an outer reforming tube wall temperature profile (T_{Wall}^{max})
 510 can be used in Eq. 4 to construct an entire profile along the reforming tube, and
 511 therefore, a specific T_{Wall}^{max} can represent a unique outer reforming tube wall tem-
 512 perature profile. The temperature dependence of $\bar{x}_{H_2}^{outlet}$ is demonstrated in Fig. 12
 513 for a wide range of outer reforming tube wall temperature profiles for which T_{Wall}^{max}
 514 varies from 1100 K to 1200 K . Due to the endothermic nature of the SMR re-
 515 actions, higher values of $\bar{x}_{H_2}^{outlet}$ are observed for temperature profiles with higher
 516 T_{Wall}^{max} .

517 In this work, all CFD reforming tube simulations under feedback control and
 518 under open-loop control are carried out in a parallel computational environment
 519 with message passing interface technology with a built-in, explicit-time-stepping,
 520 transient solver. Transient simulation may reveal dynamics of a system that cannot
 521 be obtained under steady-state simulation Pantoleonatos et al. (2012). In particular,
 522 Barton reported that the catalyst core temperatures of the water-gas shift reactors
 523 in an IGCC-TIGAS polygeneration plant can exceed the maximum steady-state
 524 value by 100 K during start-up, which would not be detected under steady-state
 525 simulation Adams and Barton (2009). Additionally, the transient solver can be
 526 used to determine the steady-state solution when a steady-state model is highly

Figure 11: The pre-determined outer reforming tube wall temperature trajectory with time for open-loop control with $\bar{x}_{H_2}^{set} = 0.465$.

527 unstable, e.g., natural convection problems with Rayleigh number in the transi-
 528 tion regime Inc. (Nov. 2013a). CFD simulations under open-loop control using
 529 a constant pre-determined outer wall temperature trajectory (e.g., Fig. 11) as the
 530 process manipulated input and built with fixed simulation settings can success-
 531 fully drive $\bar{x}_{H_2}^{outlet}$ to the desired $\bar{x}_{H_2}^{set}$ under a strictly disturbance-free environment
 532 as shown in Fig. 13. However, due to the absence of the feedback mechanism,
 533 open-loop control is unable to recognize or handle the tube-side feed disturbances,
 534 which results in a substantial deviation of the steady-state $\bar{x}_{H_2}^{outlet}$ from the desired
 535 set-point, $\bar{x}_{H_2}^{set}$. The aforementioned robustness issue motivates us to design feed-
 536 back control schemes including proportional (P), proportional-integral (PI) and
 537 optimization-based control schemes, which are utilized to produce a feedback-
 538 based manipulated input in order to drive $\bar{x}_{H_2}^{outlet}$ to the desired $\bar{x}_{H_2}^{set}$ in the presence

Figure 12: The expected steady-state value of $\bar{x}_{H_2}^{outlet}$ at different constant outer reforming tube temperature profiles which are presented by their corresponding T_{Wall}^{max} . The relationship between $\bar{x}_{H_2}^{outlet}$ and T_{Wall}^{max} is captured by the second-order polynomial, $T_{Wall}^{max} = 5648.6(\bar{x}_{H_2}^{outlet})^2 - 3814.7\bar{x}_{H_2}^{outlet} + 1699.5$, which is used to generate an appropriate manipulated input for the open-loop CFD simulations based on the desired $\bar{x}_{H_2}^{set}$ (Fig. 11).

539 of tube-side feed disturbances.

540 3.2. Feedback controller design

541 In this work, the primary objectives of the feedback control schemes are to
 542 drive $\bar{x}_{H_2}^{outlet}$ to the desired $\bar{x}_{H_2}^{set}$ in the presence of the tube-side feed disturbance,
 543 and to speed up the dynamics of the process to allow the system to quickly ad-
 544 vance to the optimized steady-state operating condition. All simulation settings of
 545 the closed-loop simulations are the same as those used in the previously studied
 546 open-loop system. It is important to emphasize that the disturbance and set-point
 547 changes are added to the process after it has already achieved a steady-state under
 548 a constant wall temperature profile.

Figure 13: The propagation of $\bar{x}_{H_2}^{outlet}$ with time under open-loop control with $\bar{x}_{H_2}^{set} = 0.465$, when the tube-side mass flow rate is kept at its nominal value (solid line), and when it is increased by 20% (dashed line). Fig. 13 demonstrates that open-loop control is not robust and cannot be used to maintain $\bar{x}_{H_2}^{set}$ at 0.465 when a change in the operating condition is imposed.

549 These feedback control schemes make the control objectives feasible and pro-
550 vide robustness against the tube-side feed disturbance, and thus resolve the criti-
551 cal drawback of open-loop control. It is desired that under each feedback control
552 scheme, the closed-loop system is initially operated in a high temperature regime
553 favoring the formation of hydrogen fuel resulting in a fast increase of $\bar{x}_{H_2}^{outlet}$ toward
554 the desired $\bar{x}_{H_2}^{set}$ due to the endothermic nature of the SMR reactions described in
555 Eq. 1. After the initial burst of hydrogen fuel, it is desired that the closed-loop
556 system is then kept in a moderately high temperature regime, which effectively
557 slows down the rapid formation of hydrogen fuel to prevent unnecessary aggres-
558 sive control action. To design feedback control schemes to accomplish the operat-
559 ing strategy, a data-driven modeling technique is used to obtain a process transfer

function describing the evolution of $\bar{x}_{H_2}^{outlet}$ based on the CFD simulation result for a step-change of the maximum outer reforming tube wall temperature from $T_{Wall}^{max}(0) = 1100\text{ K}$ to $T_{Wall}^{max}(t) = 1110\text{ K}$ (which is discussed in greater detail below). Then, this approximate model capturing the dominant dynamics of the reforming tube is combined with classical feedback control to form a closed-loop system.

It is worth noting that the process transfer function derived based on the response of $\bar{x}_{H_2}^{outlet}$ to a step-change of T_{Wall}^{max} from $T_{Wall}^{max} = 1100\text{ K}$ to $T_{Wall}^{max} = 1110\text{ K}$ was compared with other transfer functions derived based on different step-changes of T_{Wall}^{max} . Specifically, different step changes in the input, i.e., step changes in T_{Wall}^{max} from 1110 K to 1130 K , from 1130 K to 1160 K and from 1160 K to 1200 K , were considered. The variations between the transfer functions (normalized) derived from the different sets of simulation results are negligible.

The closed-loop system is utilized in a parametric study to determine the appropriate control parameters, i.e., K_c of P control or K_c and τ_I of PI control. Specifically, the initial values of the P/PI parameters are obtained by the Cohen-Coon tuning method based on the response of the process output, $\bar{x}_{H_2}^{outlet}$, to a step-change of the process input chosen as the maximum outer reforming tube wall temperature. Then the process model derived from the data-driven modeling technique based on the aforementioned simulation result is used to build closed-loop models with feedback control schemes to improve the controller parameter estimates. Finally, the P/PI parameters are incorporated into the closed-loop CFD models, and adjusted until the desired closed-loop responses are observed.

Furthermore, the maximum allowable temperature value of 1200 K is taken into account in the control schemes to avoid the rupture of the reforming tube which can occur at exceedingly high operating temperatures. Additionally, this upper limit on the outer reforming tube wall temperature prevents the deposition of carbon on the reforming tube wall and on the surfaces of catalyst particles, which would significantly decrease the heat transfer rate to the flowing gas mixture and prevent the reactants from entering the catalyst active sites resulting in a lower $\bar{x}_{H_2}^{outlet}$. Next, the minimum allowable temperature value of 987 K at the tube inlet is considered in the formulation of the control schemes to ensure that sufficient heat is transferred to the flowing tube-side gas mixture to facilitate the highly endothermic SMR reactions. Moreover, since the scope of our current work focuses on CFD modeling and controller design of the reforming tube instead of the SMR furnace, we will disregard the dynamics of the outer reforming tube wall temperature. Hence, we suppose that the outer reforming tube wall temperature can reach the predicted profile instantaneously. Lastly, we assume that measurements of $\bar{x}_{H_2}^{outlet}$ are available at all sampling instances. Classical P and PI control schemes that are based on the deviation of $\bar{x}_{H_2}^{outlet}$ from the desired set-point and account for input constraints are presented as follows:

P control scheme

$$\begin{cases} e(t_k) = \bar{x}_{H_2}^{set} - \bar{x}_{H_2}^{outlet}(t_k) \\ u_P(t_k) = K_c' e(t_k) \\ T_{Wall}^{max}(t_k + \Delta t) = \\ \quad T_{Wall}^{max}(t = 0) + u_P(t_k) \end{cases} \quad (8a)$$

PI control scheme

$$\begin{cases} e(t_k) = \bar{x}_{H_2}^{set} - \bar{x}_{H_2}^{outlet}(t_k) \\ u_{PI}(t_k) = K_c \left(e(t_k) + \frac{1}{\tau_I} \int_{t_0}^{t_k} e(\tau) d\tau \right) \\ T_{Wall}^{max}(t_k + \Delta t) = \\ \quad T_{Wall}^{max}(t = 0) + u_{PI}(t_k) \end{cases} \quad (8b)$$

$$T_{Wall}(x, t_k + \Delta t) \leq T_{max} \quad (8c)$$

$$T_{Wall}(x = 0, t_k + \Delta t) \geq T_{min} \quad (8d)$$

$$T_{Wall}(x, t_k + \Delta t) = \quad (8e)$$

$$\begin{bmatrix} x^4 & x^3 & x^2 & x^1 & x^0 \end{bmatrix} \begin{bmatrix} -0.0221 \\ 0.8003 \\ -10.734 \\ 64.416 \\ T_{Wall}^{max}(t_k + \Delta t) - 151.83 \end{bmatrix}$$

601 where t_k , t_0 and $\Delta t = 0.04$ *seconds* are the current time, the initial time, and the
 602 sampling time interval of the CFD simulation respectively, $e(t_k)$ represents the
 603 current deviation of $\bar{x}_{H_2}^{outlet}$ from the desired set-point $\bar{x}_{H_2}^{set}$, T_{min} and T_{max} are the
 604 minimum and maximum allowable reforming tube wall temperatures respectively,
 605 $K_c' = 1856.3$ and $u_P(t_k)$ are the controller gain and current controller output of
 606 the P control scheme, $K_c = 1856.3$, $\tau_I = 46.4$ and $u_{PI}(t_k)$ are the controller gain,

607 controller time constant and current controller output of the PI control scheme,
 608 $T_{Wall}^{max}(t_k + \Delta t)$ and $T_{Wall}^{max}(t = 0)$ are the predicted (i.e., prior to being saturated
 609 with the temperature constraints) and initial maximum outer reforming tube wall
 610 temperatures, respectively, and $T_{Wall}(x, t_k + \Delta t)$ is the predicted outer reform-
 611 ing tube wall temperature profile. At the end of each sampling time interval, the
 612 measurement of $\bar{x}_{H_2}^{outlet}$ is acquired from the CFD simulation, and the current devi-
 613 ation from the desired $\bar{x}_{H_2}^{set}$ ($e(t_k)$) is computed. Then, the control output ($u_{PI}(t_k)$
 614 or $u_P(t_k)$) is evaluated depending on the implemented control scheme, which al-
 615 lows $T_{Wall}^{max}(t_k + \Delta t)$ to be estimated. The value of $T_{Wall}^{max}(t_k + \Delta t)$ is subjected to
 616 Eq. 8c; if the constraint is not satisfied, $T_{Wall}^{max}(t_k + \Delta t)$ is set to the value of T_{max} .
 617 Then, $T_{Wall}^{max}(t_k + \Delta t)$ is used to compute the predicted wall temperature profile
 618 ($T_{Wall}(x, t_k + \Delta t)$), which is applied until the next sampling time, when the new
 619 value of $\bar{x}_{H_2}^{outlet}$ is obtained from the CFD simulation.

620 3.3. Closed-loop simulation results

621 In this section, we investigate the closed-loop performance of the P and PI
 622 controllers, which adjust the manipulated input variable chosen to be the outer
 623 reforming tube wall temperature, to drive $\bar{x}_{H_2}^{outlet}$ to the desired $\bar{x}_{H_2}^{set}$ in the absence
 624 or presence of a tube-side feed disturbance. At the end of each sampling time
 625 interval, the measurement of $\bar{x}_{H_2}^{outlet}$ is acquired from the CFD simulation, and the
 626 control action, which is formulated following the scheme described in Eq. 8, is
 627 evaluated. Next, the control action is applied to the closed-loop system until the
 628 end of the next sampling time, when the new $\bar{x}_{H_2}^{outlet}$ is received from the CFD
 629 simulation. In our work, the performance of the feedback control schemes is

630 evaluated based on two criteria: the improvement in the process dynamics and
 631 the final deviation of $\bar{x}_{H_2}^{outlet}$ from the desired $\bar{x}_{H_2}^{set}$. It is important to note that
 632 the process dynamics in this work is defined as the time duration that is needed
 633 for the $\bar{x}_{H_2}^{outlet}$ of the closed-loop system to first attain 99.99% of the desired $\bar{x}_{H_2}^{set}$.
 634 Unfortunately, due to the well-known drawback of P control, an offset of the final
 635 value of $\bar{x}_{H_2}^{outlet}$ is expected in the system under P control, and hence, the process
 636 dynamics cannot be evaluated based on the arrival of $\bar{x}_{H_2}^{outlet}$ at the desired $\bar{x}_{H_2}^{set}$. In
 637 the case of the P control scheme, the process dynamics is the time that is required
 638 for the closed-loop system to settle at its new steady-state.

639 In the case of set-point tracking control under a disturbance-free environment,
 640 the outer reforming tube wall temperature trajectory propagates differently under
 641 P control and PI control than under open-loop control (Fig. 11, Fig. 14(a) and
 642 Fig 14(b)). The open-loop control strategy adopts the outer reforming tube wall
 643 temperature profile based on the relationship between $\bar{x}_{H_2}^{outlet}$ and T_{Wall}^{max} as shown
 644 in Fig. 12 with $\bar{x}_{H_2}^{outlet} = \bar{x}_{H_2}^{set} = 0.465$. Because of the aforementioned closed-
 645 loop operating policy described in Section 3.2, the process dynamics is greatly
 646 enhanced in the closed-loop system shown in Fig. 15. In particular, it takes ~ 244
 647 seconds for $\bar{x}_{H_2}^{outlet}$ to attain its final value under open-loop control, while it takes
 648 ~ 90 seconds corresponding to an improvement of 63.1% under P control or
 649 ~ 154 seconds corresponding to an improvement of 36.9% under PI control,
 650 respectively. Nevertheless, the closed-loop system implemented with P control is
 651 unable to drive $\bar{x}_{H_2}^{outlet}$ completely to the desired $\bar{x}_{H_2}^{set}$, and the offset is estimated to
 652 be 3.18%.

653 Next, we turn our attention to the case of set-point tracking control under
 654 a tube-side feed disturbance to emphasize the importance of feedback control.
 655 In this case, a 20% increase in the tube-side inlet mass flow rate is introduced
 656 into the simulation settings to simulate the feed disturbance. In the presence of
 657 a tube-side feed disturbance, PI control and open-loop control yield significantly
 658 different temperature trajectories of the outer reforming tube wall (Fig. 11 and
 659 Fig. 16). Unlike feedback control schemes, open-loop control does not possess
 660 a self-correcting mechanism, and therefore, open-loop control fails to recognize
 661 the presence of the induced tube-side feed disturbance. As a result, open-loop
 662 control fails to drive $\bar{x}_{H_2}^{outlet}$ to the desired $\bar{x}_{H_2}^{set}$ (Fig. 17) since it blindly applies
 663 the pre-determined outer reforming tube wall temperature trajectory, which is in-
 664 dependent from $\bar{x}_{H_2}^{outlet}$. On the contrary, under PI control, $\bar{x}_{H_2}^{outlet}$ is driven to the
 665 desired $\bar{x}_{H_2}^{set}$ in the presence of a tube-side feed disturbance within ~ 308 sec-
 666 onds. This result demonstrates that the performance of PI control is superior to
 667 that of open-loop control (as well as P control) in the presence of a tube-side feed
 668 disturbance. Most importantly, the above analysis confirms that it is possible to
 669 utilize CFD software to model, design and implement feedback control schemes
 670 into a CFD model to form a closed-loop system to study disturbance compensa-
 671 tion.

672 **Remark 2.** Although the open-loop, P, and PI set-point tracking and disturbance
 673 rejection properties are widely known, the purpose of this work is to evaluate the
 674 closed-loop properties of the SMR system and, more importantly, to demonstrate
 675 that real-time classical and advanced feedback control schemes can be incorpo-

rated into CFD models, and specifically within the model of the industrial-scale SMR system. P/PI controllers are examples of real-time classical feedback control schemes, while dynamic optimization with integral feedback control (to be presented in detail in the next section) is an example of a real-time advanced feedback control scheme. The simulation results demonstrate the expected dynamic responses, which in turn provides necessary evidence to support the accuracy and effectiveness of the proposed methodology for real-time closed-loop feedback control design based on CFD modeling technology.

3.4. Integrating dynamic optimization and feedback

In previous sections, we demonstrate that we can design closed-loop CFD models under feedback control schemes in Ansys Fluent CFD software, and analyze and evaluate the performance of P control and PI control in the presence and absence of a tube-side feed disturbance. In both scenarios, PI control possesses the most enhanced process dynamics and zero-offset in the final $\bar{x}_{H_2}^{outlet}$, and therefore, it is ranked to have the best performance. Nevertheless, we strive to improve the process dynamics further, and in this regard, we would like to formulate more advanced control schemes, i.e., optimization-based control schemes, to design a closed-loop system in CFD software that drives $\bar{x}_{H_2}^{outlet}$ to the desired $\bar{x}_{H_2}^{set}$.

Optimization-based control schemes (e.g., model predictive control – MPC) have been widely used in the HyCO/SMR plants of the gas industry. Successful implementation of optimization-based control of SMR plants can allow more effective set-point tracking and disturbance rejection properties. Therefore, motivated by industrial practical considerations, we devote the remaining section to

699 the development of a dynamic optimization and integral feedback control scheme
700 (Fig. 18) which generates the outer reforming tube wall temperature profile.

701 The majority of approximate models formulated to capture the dominant dy-
702 namics of the reforming tube for the purpose of designing optimization-based
703 control schemes such as MPC in industry are data-based models. Hence, we first
704 derive a mathematical model capturing the relationship between $\bar{x}_{H_2}^{outlet}$ and the
705 outer reforming tube wall temperature by utilizing a data-driven modeling tech-
706 nique (previously mentioned in Section 3.3) and CFD simulation results. It is
707 worth emphasizing that all CFD simulation settings from the previous sections are
708 again used in this section. From the CFD simulation result of the step-change of
709 the maximum outer reforming tube wall temperature from $T_{Wall}^{max}(0) = 1100\text{ K}$ to
710 $T_{Wall}^{max}(t) = 1110\text{ K}$, the dynamic response of $\bar{x}_{H_2}^{outlet}$ exhibits a behavior that can be
711 described by a first-order transfer function indicated by the initial non-zero slope.
712 Hence, the discrete first-order single-input-single-output (SISO) model presented
713 in Eq. 9a below is formulated to represent the transfer function of the process.
714 Then, based on this first-order SISO model, the maximum likelihood estimation
715 (MLE) method Rogers and Steiglitz (1967); Kumar and Varaiya (1986) presented
716 in Eq. 9b is used to identify the appropriate model parameters with a set of outer
717 reforming tube wall temperature and $\bar{x}_{H_2}^{outlet}$ data from the above CFD simulation.
718 The discrete first order SISO model and MLE formulation are presented as fol-
719 lows:

$$y(k) = \frac{B(s)}{A(s)} u(k) + \frac{1}{A(s)} e(k) \quad (9a)$$

$$\begin{cases} A(s) = a_0 + a_1 s \\ B(s) = b_0 \end{cases}$$

$$\tau(y(k), u(k)) = \arg \max_{\theta \in \Theta} \{y(k) | u(k), \theta\} \quad (9b)$$

where $y(k)$ and $u(k)$ are the process output and process input respectively, s is an independent variable on the Laplace domain of the transfer function, k is a discrete time variable, $\tau(y(k), u(k))$ is the maximum likelihood estimator, $\theta = [a_0 \ a_1 \ b_0]$ is the parameter vector of the estimated model, and $e(k)$ is assumed to be a value of a white noise function with zero mean and a standard deviation of 1. Utilizing the aforementioned strategy, a_0 , a_1 and b_0 are estimated to be 2.188×10^{-3} , 1.000 and 1.764×10^{-5} respectively, and therefore, the approximate model that describes the dominant dynamics of the reforming tube is created.

Subsequently, this dynamic model is applied in the Matlab MPC Design Toolbox (see Table 6 for the parameters used) for the calculation of an optimal maximum outer reforming tube wall temperature trajectory, $T_{Wall}^{max,p}(t_k + \Delta t)$, based on the desired $\bar{x}_{H_2}^{set}$ and aforementioned constraints on the outer reforming tube wall temperature (Eqs. 8c-8d). Then, based on this reference manipulated input trajectory, the proposed dynamic optimization and integral feedback control scheme is formulated as follows:

$$\begin{cases} e(t_k) = \bar{x}_{H_2}^{set} - \bar{x}_{H_2}^{outlet}(t_k) \\ u_I(t_k) = \frac{1}{\tau_I} \int_{t_0}^{t_k} e(\tau) d\tau \\ T_{Wall}^{max}(t_k + \Delta t) = \\ \quad T_{Wall}^{max,p}(t_k + \Delta t) + u_I(t_k) \end{cases} \quad (10a)$$

$$T_{min} \leq T_{Wall}(x, t_k + \Delta t) \leq T_{max} \quad (10b)$$

$$T_{Wall}(x, t_k + \Delta t) = \quad (10c)$$

$$\begin{bmatrix} x^4 & x^3 & x^2 & x^1 & x^0 \end{bmatrix} \begin{bmatrix} -0.0221 \\ 0.8003 \\ -10.734 \\ 64.416 \\ T_{Wall}^{max}(t_k + \Delta t) - 151.83 \end{bmatrix}$$

735 where $T_{Wall}^{max,p}(t_k + \Delta t)$ is the reference manipulated input trajectory determined
 736 at $t_k + \Delta t$ from the dynamic optimization using the MPC algorithm in Matlab.
 737 At the end of each sampling time interval, a measurement of $\bar{x}_{H_2}^{outlet}$ acquired from
 738 CFD simulation is used to evaluate the current deviation of $\bar{x}_{H_2}^{outlet}$ from the desired
 739 $\bar{x}_{H_2}^{set}$, and the corresponding integral control action, $u_I(t_k)$, is determined. Then,
 740 $T_{Wall}^{max,p}(t_k + \Delta t)$, obtained from the strategy described in the preceding paragraph,
 741 is adjusted by $u_I(t_k)$ to yield the predicted $T_{Wall}^{max}(t_k + \Delta t)$ as the manipulated
 742 input to the closed-loop CFD model.

743 We compare the performance of the dynamic optimization and integral feed-
 744 back control scheme with those of PI feedback control and open-loop control
 745 based on the two aforementioned criteria for control performance in Sec. 3.2. It
 746 is worth mentioning that the primary role of integral control in the dynamic op-
 747 timization and integral feedback control scheme is to guarantee that $\bar{x}_{H_2}^{outlet}$ can

Table 6: Matlab MPC Design Toolbox parameters.

Parameter	Value
Input weight	0.00
Output weight	1.00
Overall estimator gain	0.5
Control step	0.1 <i>seconds</i>
Prediction horizon	2000 steps
Control horizon	20 steps

748 always attain the desired $\bar{x}_{H_2}^{set}$ even in the presence of a tube-side feed disturbance.
 749 In the case of set-point tracking control under a tube-side feed disturbance, both
 750 the dynamic optimization and integral feedback control scheme and the PI control
 751 scheme can drive the system to the desired set-point; however, the dynamic opti-
 752 mization and integral feedback control scheme predicts a slightly different outer
 753 reforming tube temperature trajectory than that predicted by PI control as shown in
 754 Fig. 20. Specifically, the maximum outer reforming tube wall temperature of the
 755 closed-loop system under the dynamic optimization with integral feedback con-
 756 trol scheme is initially maintained at 1200 *K* for nearly 40 *seconds* as shown in
 757 Fig. 19, which speeds up the process dynamic response. Based on the metric that
 758 we previously defined in this study, it only takes ~ 33 seconds for $\bar{x}_{H_2}^{outlet}$ to first
 759 achieve the desired $\bar{x}_{H_2}^{set}$ under the dynamic optimization with integral feedback
 760 control scheme as shown in Fig. 20, corresponding to an improvement of 89.3%
 761 when compared with the system under PI control for which it takes ~ 308 sec-
 762 onds. Nevertheless, the aggressive manipulated input generated by the dynamic
 763 optimization with integral feedback control scheme also causes the oscillation of

764 $\bar{x}_{H_2}^{outlet}$ around the desired $\bar{x}_{H_2}^{set}$. Therefore, it is important to notice that although the
765 dynamic optimization with integral feedback control scheme allows $\bar{x}_{H_2}^{outlet}$ to ad-
766 vance to the desired $\bar{x}_{H_2}^{set}$ much faster than PI control, it ultimately requires a longer
767 duration to reach the optimized steady-state conditions and results in oscillation;
768 therefore, its performance is comparable with that of PI feedback control.

769 **Remark 3.** As discussed above, the PI controller is easier to implement than the
770 dynamic optimization with integral feedback control scheme but it does not use
771 an explicit plant model, and as a result, the closed-loop response is slower be-
772 cause the tuning was chosen to avoid high sensitivity to disturbances. Further, the
773 overall PI closed-loop response was not improved for other tunings investigated
774 that reasonably avoided this sensitivity (another result is discussed below that con-
775 firms the sufficiency of the PI controller with the chosen tuning by demonstrating
776 its robustness to various disturbances). However, the dynamic optimization with
777 integral feedback control scheme achieves a faster closed-loop response than the
778 PI controller with the knowledge of the process dynamics; plant-model mismatch,
779 integral action, and the effect of the disturbance may contribute to the oscillations
780 in the output trajectory. The comparison results demonstrated that both control
781 schemes force the output to track the set-point and deal well with disturbances.
782 As noted, the sufficiency of the chosen PI controller tuning is further validated
783 by testing the closed-loop response of $\bar{x}_{H_2}^{outlet}$ to a different disturbance than that
784 utilized in the results presented above. An additional disturbance in the tube-
785 side feed, i.e., the temperature of the tube-side feed is increased by 10% from its
786 nominal value, is induced to validate the robustness of the PI controller against

different potential disturbances. The simulation results including the control action (Fig. 21) and corresponding process response (Fig. 22) generated by the PI controller to counteract the tube-side feed temperature disturbance are similar to those of the previous case in Sec. 3.3 when the induced disturbance is the tube-side feed mass flow rate. Specifically, $\bar{x}_{H_2}^{outlet}$ is successfully driven to the desired $\bar{x}_{H_2}^{set}$ in the presence of a tube-side feed temperature disturbance within ~ 200 seconds.

4. Conclusions

This work initially focused on demonstrating that CFD software can be employed to create a detailed CFD model of an industrial-scale steam methane reforming tube, and subsequently focused on the design and implementation of feedback control schemes into the CFD model. The simulation results of the reforming tube CFD model simulating the transport and chemical reaction phenomena with an approximate representation of the catalyst packing matched well with the available industrial plant data. The closed-loop CFD simulation results demonstrated that the proposed PI control scheme and a control scheme combining dynamic optimization and integral feedback control could drive the value of $\bar{x}_{H_2}^{outlet}$ to a desired $\bar{x}_{H_2}^{set}$, and significantly improve the process dynamics compared to that under open-loop control. In the case of set-point tracking control under a tube-side feed disturbance, the dynamic optimization with integral feedback control scheme calculates a more aggressive outer reforming tube wall temperature trajectory compared to that calculated by PI control but both control systems drive

809 the average outlet hydrogen mole fraction to the set-point.

810 **5. Acknowledgements**

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(a)

(b)

Figure 14: The outer reforming tube wall temperature profile trajectory in the absence of a tube-side feed disturbance under P control shown in Fig. 14(a) and under PI control shown in Fig. 14(b) with $\bar{x}_{H_2}^{set} = 0.465$.

Figure 15: The propagation of $\bar{x}_{H_2}^{outlet}$ with time in the absence of a tube-side feed disturbance under P control (solid line) and under PI control (dashed line). The open-loop system response (dashed-dotted line) is also included for a wall temperature profile for which $\bar{x}_{H_2}^{set} = 0.465$.

Figure 16: The outer reforming tube wall temperature profile trajectory in the presence of a disturbance in the tube-side feed (20% increase in tube-side mass flow rate) under PI control with $\bar{x}_{H_2}^{set} = 0.465$.

Figure 17: The propagation of $\bar{x}_{H_2}^{outlet}$ with time in the presence of a disturbance in the tube-side feed (20% increase in tube-side mass flow rate) under PI control (solid line) and under open-loop control (dashed line) for a wall temperature profile for which $\bar{x}_{H_2}^{set} = 0.465$.

Figure 18: Dynamic optimization and integral feedback control scheme, where the manipulated input at $t_k + \Delta t$ is computed based on the reference manipulated input profile at $t_k + \Delta t$ and the integral control action at t_k , which is described in Eq. 10.

Figure 19: The outer reforming tube wall temperature profile trajectory in the presence of a tube-side feed disturbance (20% increase in tube-side mass flow rate) under the dynamic optimization with integral feedback control scheme with $\bar{x}_{H_2}^{set} = 0.465$.

Figure 20: The propagation of $\bar{x}_{H_2}^{outlet}$ with time in the presence of a tube-side feed disturbance (20% increase in tube-side mass flow rate) under the dynamic optimization with integral feedback control scheme (solid line) and under PI control (dashed line). The open-loop system response (dashed-dotted line) is also included for a wall temperature profile for which $\bar{x}_{H_2}^{set} = 0.465$.

Figure 21: The outer reforming tube wall temperature profile trajectory in the presence of a disturbance in the tube-side feed temperature under PI control with $\bar{x}_{H_2}^{set} = 0.465$.

Figure 22: The propagation of $\bar{x}_{H_2}^{outlet}$ with time in the presence of a disturbance in the tube-side feed temperature (solid line) and in the presence of a disturbance in the tube-side feed mass flow rate (dashed line) under PI control for a wall temperature profile for which $\bar{x}_{H_2}^{set} = 0.465$.