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## Technical Challenges in Fuel Flexible SOFC/GT Hybrid Systems: Coupling Effects of Cathode Air Mass Flow

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#### 8

#### 9 Abstract

10 Considering the limited turndown potential of gasification technologies, supplementing a fuel cell turbine 11 hybrid power system with natural gas provides flexibility that could improve economic viability. The dynamic characterization of fuel composition transients is an essential first step in completing the system identification 12 13 required for controls development. In this work, both open loop and closed loop transient responses of the fuel cell in a solid oxide fuel cell (SOFC) gas turbine (GT) hybrid system to fuel composition changes were 14 15 experimentally investigated using a cyber-physical fuel cell system. A transition from coal-derived syngas to methane rich gases with no turbine speed control was studied. The distributed performance of the fuel cell was 16 17 analyzed in detail with temporal and spatial resolution across the cell.

Dramatic changes in fuel cell system post combustor thermal output or "thermal effluent" resulting from anode composition changes drove turbine transients that caused significant cathode airflow fluctuations, by as much as 8% in less than a minute. In comparing the open loop responses to identical tests conducted under closed loop conditions without significant airflow changes, it was discovered that the cathode airflow change was a major linking event in short-term system transient response. The results suggested that modulating cathode air flow in response to fuel composition changes offers promise for the dynamic control of SOFC/GT hybrid systems with fuel flexibility.

Keywords: Open loop characterization; fuel composition changes; cathode air mass flow; fuel cell gas turbine hybrid; cyber-physical simulations.

#### 28 1. Introduction

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30 High temperature solid oxide fuel cells (SOFC) are most beneficial for power generation hybridization 31 with thermal-based generator systems as compared to other fuel cell technologies because of their high operating 32 temperatures. For example, SOFCs can be integrated with a bottoming gas turbine cycle to exploit the benefits of 33 high quality waste heat and fuel recovery from the fuel cell stacks for additional power production [1, 2]. Directfired solid oxide fuel cell gas turbine (SOFC/GT) hybrid systems shown in Figure 1 also offer some advantages 34 35 in terms of air pressurization as a result of turbomachinery pressure ratio effect on fuel cell Nernst potentials, and 36 heat recovery of gas turbine exhaust [3]. Both high pressure and heat recuperation directly help the total system 37 efficiency [3]. SOFC/GT systems are also considered economically feasible for early technology adoption because 38 the fuel cell lifetime could be extended in a hybrid configuration [4]. Although a commercial direct-fired 39 SOFC/GT plant does not exist yet, the U.S. Department of Energy (DOE), National Energy Technology 40 Laboratory (NETL) has built a world-leading cyber-physical system that mimics an SOFC system in a hybrid configuration using the seamless integration of a numeric model with hardware, and then coupled the hardware 41 42 to a real recuperated gas turbine cycle [5]. The hardware test facility at NETL is able to emulate SOFC/GT hybrid 43 dynamics performance, aimed at improving the system flexibility and achieving DOE efficiency targets.

44 The potential of fuel flexibility in high temperature SOFCs makes this technology more feasible to handle 45 fuel composition fluctuations or drastic changes in fuel type. At an operating temperature higher than 600°C, 46 SOFCs have higher potential to run on various conventional fuels (e.g. natural gas, coal-derived syngas, and 47 reformed diesel) and alternative fuels (e.g. biogas, ethanol, and biodiesel). An optimization study in advanced 48 power generation systems that considered both economics and environmental impacts has suggested that flexible 49 operations are important for meeting new economic situations that experience changes in fuel prices or new energy 50 policies, and current as well as expected environmental regulations [5]. The study showed that the net present 51 value (NPV) of a polygeneration plant for power and chemical production could improve up to 63% if the system 52 is 100% flexible [6]. In this example, dynamic shifting in fuels can be one possible strategy to manage fuel cell 53 and gas turbine power for load following or chemical production balance during peak seasons and off-peak 54 seasons to maximize the plant profits.

55 However, some practical issues may prevent changes to fuel composition input to the existing SOFC 56 technologies because different fuels result in different temperature performance, fuel utilization, and

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57 electrochemical properties, all of which play an important role in the lifetime of a fuel cell [7, 8]. Thermal 58 management was identified to be one of the greatest challenges to operate SOFC systems using different fuel 59 compositions or fuel types [7, 9]. For instance, the use of methane in SOFCs could cause problematic temperature 60 gradients across the cell due to the endothermic cooling effect of internal methane reforming. Disproportionate cooling and heating may also occur locally along the cell as a result of internal methane reforming, water-gas 61 shifting, and electrochemical reactions. Thus, fuel cell performance degradation will be a major hurdle since the 62 63 degradation rate may be accelerated if the systems demonstrate lower temperature profiles or high current density 64 and high fuel utilization [10]. Lowering the fuel cell stack temperature in SOFC/GT hybrids will decrease the turbine speed and turbine efficiency, which eventually affect the upstream conditions of SOFC stacks [11-13]. 65



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Figure 1: Basic flow diagram of a SOFC/GT hybrid system

70 Considering the strong coupling between fuel cell systems and the balance of the SOFC/GT plant, 71 problems with system performance as a result of fuel flexible operations could be arise. Severe fuel cell damage 72 due to thermal stress, unbalanced pressure between anode and cathode, shaft over speed, compressor surge and 73 stall may result if the transient is too fast [14-16]. In fact, controls for thermal management and degradation for 74 flexible fuel operations are not available yet. In addition, the range of fuel flexibility that could be implemented 75 in SOFC systems is still under research [17]. Although the aforementioned challenges during fuel dynamic 76 operations could be expected based on simulation results or available observations in standalone fuel cell systems, 77 the consequences in the hybrid cycles have not yet been investigated experimentally. Without experimental testing 78 on a hardware system, modeling and numerical simulation efforts are still insufficient to capture and understand 79 the SOFC/GT transient response and operational challenges that may help in developing control strategies.

80 Previous studies considered hybridizing gasifier technologies for biomass, coal-derived syngas, and 81 liquid fuels to exploit the SOFC fuel flexibility benefits [1, 2]. The influences of fuel gas compositions on the 82 total system efficiency, economic, and thermodynamic feasibility in a specific SOFC hybrid system design are 83 among active research topics in most studies [1, 18-20]. Previous work has also considered the consequences of 84 internal reforming systems and external reforming systems, which were driven by the use of natural gas or 85 methane [21]. However, the studies on fuel flexibility of SOFC/GT hybrid systems were all conducted via 86 modeling and numerical simulations at steady state conditions. It is clear that transient performance of SOFC/GT 87 and controls development are much less commonly studied, primarily due to the lack of available experimental 88 data [22, 23].

In this paper, the experimental evidence of SOFC/GT dynamic performance in response to fuel 89 90 composition transients is presented, followed by a characterization of transient trajectory for each key process 91 variable. First, we ran the test without using any controller in the system such that the results could provide 92 understanding of the actual transient impact on fuel cell temperature. Second, we repeated the same test with a 93 turbine speed controller to compare and investigate the influence of cathode air mass flow during fuel composition 94 dynamics. We found that the effects on fuel cell temperature mainly drove the dynamics coupling between all 95 SOFC/GT hybrid components and the resulting cathode air mass flow fluctuation could be the linking event in 96 the initial dynamic response. The findings presented in this paper were uniquely collected using hardware-based 97 simulations of an SOFC/GT to support future development of dynamic controls for SOFC/GT thermal 98 management under fuel flexible environments. 99

100

#### 102 2. Background and Methodology

Many researchers selected cathode air mass flow as a critical manipulated variable for SOFC thermal 104 105 management in hybrid systems because the impact of cathode air mass flow on temperature dynamics was 106 significantly strong and fast [7, 24]. Cathode air mass flow control was used for various objectives, including 107 during start-up, power output control [25, 26], load following [27, 28], or part load operations [29, 30]. Cathode 108 air mass flow was manipulated to minimize spatial temperature variation and to maintain the average stack 109 temperature such that thermal stress in the cell can be avoided [14, 24, 31]. However, fuel composition effects 110 were not considered in the thermal management and control development due to limited studies in fuel 111 composition transients.

Motivated by the unknown transient behaviors of the SOFC/GT hybrid systems under fuel dynamics, we started investigating the impacts of a step change from coal-derived syngas to 14 mol% CH<sub>4</sub> and 86% steam in a closed loop experiment [17]. The turbine speed was maintained constant to have a constant cathode air mass flow [9, 17]. This base case was beneficial to test the system within very minimal disturbance and failure risk since the actual response was unknown. The turbine speed control also allowed decoupling the effects of cathode air mass flow variation resulted from the fuel composition dynamics.

118 Although the main scope of this paper concern open loop system characterization, closed loop tests were 119 performed as well for comparison purposes. In this work, the step tests were slightly different from the previous 120 work; in this work the feed composition was given a step change from coal-derived syngas to 13.6 mol% methane 121 and 86.4% steam. This is because scoping studies using 14 mol% methane and 86% steam in open loop (with no 122 turbine speed controller) led to dangerous circumstances and premature stoppage since the turbine quickly 123 approached the turbine speed safety limit [32]. However, lowering the target methane content by only 3% to a 124 composition of 13.6 mol% CH<sub>4</sub>, 86.4% steam resulted in a stable turbine speed in open loop with no safety violations. Therefore, in this work, none of the results of the previous work could be used for comparison purposes, 125 126 and so both open loop and closed loop studies using 13.6 mol% methane step targets were conducted for this 127 work.

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#### 129 2.1. Description of Cyber-physical Simulation System

130 The U.S Department of Energy, National Energy Technology Laboratory (NETL) in Morgantown, West Virginia developed a Hybrid Performance (Hyper) project test facility to characterize SOFC/GT hybrid systems 131 132 for dynamic control development using a cyber-physical fuel cell system. Hyper, as shown in Figure 2, was used 133 to investigate system transient capabilities that are associated with feasible dynamic operating ranges, coupling effects between fuel cell subsystem and recuperated gas turbine cycle, and highly complex dynamic control 134 135 strategies [15, 16, 33-35]. In the Hyper project, a previously developed real-time dynamic model (in the dotted 136 box in Figure 2) was coupled to real hardware to emulate SOFC dynamic performance. We used a one-137 dimensional (1D) dynamic model that was able to simulate the SOFC every 30 ms [36]. This cyber-physical fuel 138 cell system was then hybridized with a real recuperated gas turbine cycle in a direct-fired configuration [5, 36]. 139 The cathode stream was physically provided in the hardware system using the compressor flow, and the anode 140 fuel stream was simulated in the model. Both hardware and software were well-integrated for real time hardware-141 based simulations to closely represent SOFC/GT hybrid transients for controls development.

142 The real-time SOFC model used in this study was developed based on a planar design and co-flow fuel 143 cell configuration. This numerical model characterized the fuel performance on a distributed basis with respect to 144 space in the direction of fuel and oxidant flow [36]. Fuel constituents fed into the system model could be a combination of CH<sub>4</sub>, CO, CO<sub>2</sub>, H<sub>2</sub>, H<sub>2</sub>O, and N<sub>2</sub>, assuming hydrogen was the only electrochemically active 145 146 component for hydrogen oxidation, as expressed in Eq. 1 [37]. The direct internal reforming reaction of methane 147 and water-gas shift reaction for carbon monoxide were considered in the model, according to Equations 2 and 3. 148 Considering the faster kinetics of the water-gas shift reaction compared to methane reforming kinetics at the 149 temperature range used in this study, the shifting reaction was assumed to occur at equilibrium [37].

151 Hydrogen electrochemical oxidation:

152  $H_2 + 0^= \to H_2 0 + e^= (\Delta H^\circ = -286 \, kJ/mol)$  (1) 153

154 Water-gas shifting:

| 155 | $CO + H_2O \rightleftharpoons H_2 + CO_2$ | $(\Delta H^{\circ} = -41 \ kJ/mol)$ | (2) |
|-----|-------------------------------------------|-------------------------------------|-----|
| 156 |                                           |                                     |     |

157 Steam methane reforming:

158 
$$CH_4 + H_2 0 \rightleftharpoons 3H_2 + CO \ (\Delta H^\circ = 206 \, kJ/mol)$$
 (3)

159

In general, the model incorporated dynamic calculation for thermal performance (heat generation, solid and gas temperature profiles), electrochemical characterization (Nernst potential, polarization losses, current 162 density, and voltage), anode composition gradients, and associated fuel cell variables (power, fuel cell post 163 combustor thermal effluent, etc.). Finite difference and finite volume were respectively used for the resolution of thermal profiles, and electrochemical performance. A detailed work of the model development was published 164 165 elsewhere [36].

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Figure 2: The layout of SOFC/GT testing facility at NETL [17].

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- 172 2.2. Open Loop and Closed Loop Test Procedures

In this study, the standard startup procedures established by NETL researchers was used to bring the 173 174 hybrid system to a steady state for cyber physical simulations [38]. Coal-derived syngas was fed as the SOFC fuel 175 at 145 g/s. The corresponding initial fuel utilization was approximately 67%. A step change in SOFC fuel 176 composition from the coal-derived syngas feed to methane-rich gases was simulated once the system was in steady 177 state. The methane-rich fuel contained 13.6 mol% CH<sub>4</sub> and 86.4% steam, which was sufficient to avoid carbon 178 deposition in the cell [7, 39].

179 Fuel flow (145 g/s) and SOFC load (220 A) were held constant over the course of the experiment to 180 investigate the impacts of fuel composition uniquely, without confounding the data with the dynamic impacts of 181 fuel flow and SOFC load. In contrast, overall fuel utilization was the result in this study that changed accordingly to fuel composition gradients. The fuel switch was simplified by changing the feed composition to the SOFC 182 183 model instantaneously, not taking into account the lag in the fuel manifold process.

184 Such a fuel transition caused a 20% decrease in overall heat input (LHV) of the anode feed. This change 185 was feasible for open loop tests in which turbine speed control was not implemented [17, 32]. Previous scoping 186 studies suggested that the coal-derived syngas had to be switched to a lower heating value methane rich fuel in 187 order to avoid turbomachinery stall and surge, excessive anode-cathode pressure differences, and cathode inlet 188 temperature excursions resulting in adverse impacts on the functionality of SOFC/GT hardware facility [17].

189 For the purposes of comparison, the test procedures and conditions used for the open loop tests and the closed loop tests in this work were identical, including fuel compositions, initial conditions of SOFC/GT hybrid 190 191 test facility, cell geometry, and hardware operation techniques. However, unlike the closed loop studies where a control system placed a modulated electrical load (or resistive turbine load) on the turbine shaft to maintain 192 193 constant turbine speed at 40,500 rpm, no turbine speed control scheme was used in the open loop tests. Thus, the 194 turbine responded directly to the total SOFC system post combustor thermal output (i.e. fuel cell thermal effluent). 195 The heat was delivered to the turbine inlet in real time as shown in Figure 2.

In this work, the 1D real-time SOFC model calculated the SOFC waste heat at every 80 ms simulation time step, considering inlet conditions of the cathode stream in the plant, user-defined inputs, and fuel cell geometry described in Table 1 [36,40]. In our cyber-physical simulations, the resulting waste heat was used to control fuel valve position of FV432 in Figure 2 in feed-forward mode. The fuel valve opened accordingly to deliver an equivalent natural gas flow to simulate the waste heat delivered to the real gas turbine cycle.

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Table 1 SOFC parameters and initial operating conditions

| System parameter                  |                                                                     |  |  |
|-----------------------------------|---------------------------------------------------------------------|--|--|
| Fuel cell load                    | 220 A                                                               |  |  |
| Anode recycle                     | 0%                                                                  |  |  |
| Initial fuel cell temperature     | 800°C                                                               |  |  |
| Total cell area                   | 200mm x 200mm                                                       |  |  |
| Anode thickness                   | 0.5mm                                                               |  |  |
| Electrolyte thickness             | 0.008mm                                                             |  |  |
| Cathode thickness                 | 0.05mm                                                              |  |  |
| Oxidant/fuel channel size         | 2mm x 2mm                                                           |  |  |
| Stack size                        | 2500 cells                                                          |  |  |
| Total stack mass                  | 3500kg                                                              |  |  |
| Total stack heat capacity         | 2625 kJ/K                                                           |  |  |
| Fuel cell cathode inlet condition |                                                                     |  |  |
| Air mass flow rate                | 1.03 kg/s                                                           |  |  |
| Air temperature                   | 705°C                                                               |  |  |
| Air pressure                      | 347 kPa                                                             |  |  |
| Air composition                   | 21% O <sub>2</sub> , 79% N <sub>2</sub>                             |  |  |
| Fuel cell anode inlet condition   |                                                                     |  |  |
| Fuel mass flow rate               | 145 g/s                                                             |  |  |
| Fuel temperature                  | 800°C                                                               |  |  |
| Fuel pressure                     | 347 kPa                                                             |  |  |
| Initial composition (mol          | CH <sub>4</sub> 0%, CO <sub>2</sub> 12.0%, CO 28.6%, H <sub>2</sub> |  |  |
| fraction)                         | 29.1%, H <sub>2</sub> O 27.1%, N <sub>2</sub> 3.2%                  |  |  |
| Final composition                 | CH <sub>4</sub> 13.6%/H <sub>2</sub> O 86.4%                        |  |  |
| Fuel cell initial condition       |                                                                     |  |  |
| Cell voltage                      | 0.83V                                                               |  |  |
| Fuel utilization                  | 67%                                                                 |  |  |

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#### 207 **3. Results and Discussion**

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#### 209 3.1. Turbine Speed and Cathode Air Mass Flow Transient Responses

The trajectories of turbine speed and cathode inlet mass flow for the open loop and the closed loop tests in response to fuel composition changes are shown in Figure 3. The time at which the step change in fuel composition occurred is indicated as time t=0. For the open loop case, the turbine speed varied depending on the changes in fuel cell thermal effluent.

215 As shown in Figure 3a, a 4% increase in turbine speed immediately after the step change in fuel 216 composition caused an 8% initial transient increase in cathode air mass flow as presented in Figure 3b. This was followed by a steady decrease to a new steady state with the turbine speed 6% lower and the cathode air flow rate 217 218 12% lower than it was at the initial condition. Figure 3c shows that the cathode inlet air mass flow was linearly 219 correlated to the turbine speed with a factor of 2:1 (R<sup>2</sup>=0.993 on a normalized comparison of the measurements) 220 at any given time. For comparison, the results from the closed loop test for the same transition is also shown in 221 Figure 3. In the closed loop test, the turbine speed was controlled at a constant 40,500 rpm. The "crossover point," 222 the point at which the open loop and the closed loop values were the same, for both variables is at around 1,600 s 223 after the step change. In this paper, crossover point was used to characterize the influence of open and closed loop 224 operations on the transient response. Delay in crossover points were investigated to identify the coupling effects 225 between all SOFC/GT process variables, which is critical developing control strategies for non-linear responses.



Figure 3: Turbine speed and cathode air mass flow responses for open loop and closed loop

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229 3.2. Transient Characterization of Fuel Cell Thermal Performance 230

#### 231 Fuel Cell Solid Temperature

232 The transients of the fuel cell solid temperature are shown in Figure 4a and 4b. In general, the fuel cell 233 solid temperatures for both the open loop (Figure 4a) and closed loop (Figure 4b) increased across the fuel cell 234 length. However, the temperature reduced with time to a lower final average temperature as the test continued 235 over the course of 5,500 s. In the open loop test, the temperature difference between the cell inlet and the cell exit 236 was 125°C at the initial steady state, before the fuel composition change and also at the final steady state after the 237 fuel composition change. The same inlet to outlet temperature difference across the cell was observed in both 238 cases, but a lower average temperature at the final steady state was realized due to the higher airflow in the closed 239 loop case.





Figure 4: Fuel cell solid temperature as a function of time, (a) Open loop (b) Closed loop

Figure 5a shows the distributed profiles of solid temperature at certain times, resulting from the open loop and closed loop tests. Figure 5b highlights the magnitude of difference between the open loop and closed loop solid temperature profiles over 5,500 s,  $(T_{solid_{CL}} - T_{solid_{OL}})$ , for selected nodes.

247 Although both the open loop and closed loop tests started at the same initial states, indicated by the overlapping profile at 1 s in Figure 5a, the solid temperature in the closed loop test was generally higher than in 248 249 the open loop over the first 2,450 s. The greatest temperature difference between the open loop and closed loop 250 occurred at node 2 within the first 650 s (Figure 5b) because solid temperature in the open loop test dropped 251 substantially. This transient was driven by rapid increase in turbine speed in the open loop case that increased 252 cathode air mass flow to the maximum value, on the order of few seconds after the fuel composition changes. 253 Therefore, the fuel cell solid was further cooled in the open loop case, in addition to direct influence of steam 254 methane reforming. As a result, the temperature decreased faster than in the closed loop case, which was more 255 evident at the beginning of the cell. This effect was shown by the increasing temperature gaps between the open 256 loop and closed loop cases up to 1,500 s.

257 As indicated in Figure 5b, the location of the greatest temperature difference shifted further from node 2 258 to node 10, at point A. At this time, the decrease in the open loop solid temperature half way down the fuel cell 259 length became more significant even though turbine speed and cathode air mass flow shown in Figure 3 started 260 to reduce from the maximum values. Decreasing cathode air mass flow should have mitigated fuel cell solid 261 temperature reduction. However, the cooling effects were still significant because the total cathode air mass flow 262 in the open loop system was still higher than in the closed loop, and the temperature dynamics were slow. The 263 maximum temperature difference of 17°C (point B) was observed at node 10 at 1,300 s after the fuel composition 264 change. After the crossover in turbine speed at 1,600 s, the reduction in fuel cell solid temperature continued 265 slowly with the subsequent cathode air mass flow reduction until the temperature crossover point was reached. 266



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Figure 5: Comparison of fuel cell solid temperature performance, between the open loop and the closed loop tests (a) Distributed data as a function of time, and (b) The response of solid temperature difference at selected fuel cell nodes

272 It is shown in Figures 5a and 5b that crossover points of solid temperature at the beginning of the cell 273 were achieved at 2,450 s after the step change, whereas the crossover points for the end of the cell were delayed 274 12 minutes later than the cell entrance. Such a significant difference in crossover points between the cell entrance 275 and the cell exit was dominated by the resulting temperature profiles from the open loop and closed loop tests, 276 indicating coupling between the temperature reduction due to convective heat transfer and methane reforming 277 kinetics. In general, the increased cathode air mass flow before the crossover point of turbine speed caused the 278 decreased in solid temperature, which finally decelerated the reforming reaction. As a result, this modified the 279 distribution of methane in the fuel cell, which in turn, affected the solid temperature.

The comparison shown in Figure 5a demonstrated a greater drop in the closed loop solid temperature after the crossover points. The solid temperature in the closed loop test kept decreasing until 7,000 s after the fuel composition switch, at which the test was terminated. Unlike the open loop process, the constant cathode air mass flow provided by the constant turbine speed operation in closed loop continued to propagate the reduction in fuel cell solid temperature throughout the test. This was clearly reflected by cathode inlet temperature profiles shown in Figure 6.





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Figure 6: Cathode inlet temperature profiles for the open loop test using 13.6 mol% CH<sub>4</sub> and for the closed loop operations using 13.6 mol% CH<sub>4</sub> and 14 mol% CH<sub>4</sub>

In contrast, the open loop temperature did not change significantly after 3,000 s from the start of the test because cathode air mass flow decreased continuously after the crossover point. The operation with variable turbine speed consequently brought the system close to a thermal equilibrium state faster than the closed loop system. Because of the variations in cathode air mass flow, the system demonstrated two distinct and opposite coupling trends in the short-term period and the long-term period. This non-linear response of temperature to composition changes suggests that complex controls in thermal management are required to take advantage of the inherent flexibility of hybrid systems.

#### 300 Spatial Gradient of Fuel Cell Solid Temperature

The impact of fuel composition changes on fuel cell solid temperature was further characterized by quantifying the spatial temperature gradient (dT/dx) along the cell, as indicated in Figure 7. Note that the cell temperature distribution should be maintained as constant as possible such that the temperature gradient could be minimized to prevent thermal stress in fuel cell components. The distribution of the temperature gradient for 20 nodes resulting from the open loop test is shown in Figure 7a, while the maximum spatial temperature gradients at critical nodes for open loop and closed loop tests are presented in Figure 7b.





Figure 7: Spatial temperature gradient as a function of time, (a) Open loop transient response (b) Comparison of open loop and closed loop solid temperature gradients at critical nodes

310 311 Both tests demonstrated the same variations and locations of the maximum dT/dx, as simplified in Figure 7b. However, the maximum dT/dx resulting from the open loop test were slightly higher than in the closed 312 loop. Before fuel composition was switched in the open loop operation, the highest temperature gradient of 11.7 313 K/cm occurred at node 5, 25% of the total fuel cell length, and gradually shifted to the next node towards the cell 314 315 exit after initiation of the transient. The greatest dT/dx of 17 K/cm appeared at node 7, 35% of the way down from the inlet, as soon as 500 s after switching to humidified methane. This value exceeded the limit of dT/dx. 316 317 Thus, adequate thermal management was required since there was high risk of thermal stress due to the excessive 318 dT/dx, which could also lead to high degradation rate impact. The maximum dT/dx was settled at node 13, 65% of the way down the length from the cell entrance, at approximately 1,950 s after the transient where cathode air 319 320 mass flow was 4% below its initial value (Figure 3b).

As shown in Figure 8, the maximum magnitude of discrepancy in the dT/dx between the open loop and closed loop at node 5 and node 7, were about 1 K/cm at 1,600 s, and 1 K/cm difference at node 13, at 650 s after the transient. As presented in Figure 9, the increasing difference in dT/dx for all three nodes were evident within the first 650 s (region I), which respectively collocated with increasing cathode air flow, before the crossover point. Note that cathode air flow in the open loop test started to decrease in region II approaching the initial condition and continued to reduce more significantly below the initial condition in region III.

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Figure 8: The difference in solid temperature gradient at critical nodes

#### 331 *Cathode Gas Temperature*

332 The dynamic transients of the fuel cell gas temperature are illustrated in Figure 9. As clearly shown in 333 Figure 9b, there was significant deviation in fuel cell gas temperature at the beginning of the cell between the 334 open loop and closed loop profiles at 250 s. This transient was attributed to the combined effects of cathode air 335 mass flow and cathode inlet air temperature. Both tests demonstrated the initial transient rise in cathode inlet air 336 temperature in response to fuel composition changes (Figure 6). However, the open loop performance shown in 337 Figure 6 experienced 1% increase, while the closed loop case experienced 3% increase. This difference in 338 temperature increase existed in spite of the higher turbine inlet temperature of the open loop operation because 339 turbine efficiency in the open loop improved at the higher speeds. As such, turbine exhaust temperature in open 340 loop case was lower because the increase in turbine exhaust temperature was not as significant as the increase in 341 turbine inlet temperature. As a consequence of the lower cathode air mass flow with higher cathode inlet air 342 temperature, gas temperature of the closed loop test at the cell entrance was higher than the open loop system at 343 250 s. 344

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Figure 10 shows the fuel cell gas temperature difference at the beginning, center, and exit of the cell. Over 5,500 s of test, the magnitude of gas temperature difference across the fuel cell length varied significantly due to fluctuations of cathode air mass flow in the open loop case. The long term trends of gas temperature started to twist after the crossover points, similar to the solid temperature dynamics shown in Figure 5. In a longer experimental run, the gas temperature in the closed loop system reduced faster than in the open loop case due to decreased solid temperature and constant cathode air mass flow at a higher level. In contrast, the open loop gas 356 temperature (Figure 9b) changed slowly as cathode air mass flow decreased, leading to insignificant temperature

357 drop between 3,000 s and 5,500 s.





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361 Figure 10: Crossover points in fuel cell gas temperature difference between the open loop and the closed loop 362 for selected local fuel cell positions

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#### 364 Cathode Solid and Gas Temperature Difference and Heat Flux

Figure 11 shows the resulting temperature difference between fuel cell solid and gas temperature 365  $(T_{solid} - T_{gas})$ . As shown in Figure 11, the temperature difference at the first 5 nodes reduced drastically at the 366 initial transients and fluctuated more significantly over the course of the test as compared to the remaining fuel 367 368 cell length. The temperature difference at the cell inlet shown in Figure 11b dramatically reduced from 76°C to 369 slightly lower than -20°C within 5,500 s, whereas the temperature difference at the end of the cell was maintained 370 fairly constant.



372 Figure 11: Fuel cell solid-gas temperature difference as a function of time, (a) Open loop transient response, (b) 373 Comparison of open loop and closed loop transients

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$$386 \qquad \dot{q} = \frac{\kappa_g N u}{D} \left( T_{solid} - T_{gas} \right) \tag{4}$$

where  $k_g$  is the thermal conductivity of gas, Nu is the Nusselt number, D is the hydraulic diameter of the gas channel,  $T_{solid}$  is the solid temperature, and  $T_{qas}$  is the gas temperature.

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Figure 12: Heat flux of a fuel cell stack as a function of time, (a) Open loop transient response, (b) Comparison
 of open loop and closed loop transients

394 The dynamic transients of heat flux at 5,500 s contrasted the initial transient trends. Because of the reversed trends of cathode air mass flow and cathode inlet air temperature after the crossover points, the heat flux 395 in the open loop test was more negative than in the closed loop test. The open loop gas temperature increased 396 397 faster than in the solid temperature such that the solid-gas temperature difference reduced, due to decreasing 398 cathode air mass flow and higher cathode inlet air temperature. In fact, as shown in Figure 11b, the gas temperature 399 at 5,500 s was higher than the solid temperature. Hence, the direction of heat flux in the long-term operation 400 changed from the solid-to-air flow to air flow-to-solid. The air flow began to heat the fuel cell system after 470 s. 401 In general, the magnitude of heat flux in the closed loop case was much lower than the open loop profiles 402 throughout the test despite of the same performance and heating impacts at the beginning of the cell.

403 At 5,500 s, the beginning of the cell was heated by the airflow, while the remainder of the fuel cell was 404 cooled by the airflow. This was a consequence of decreasing fuel cell solid temperature toward the end of the test 405 and the methane reformation region shifting further down the fuel cell length.





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Figure 13: Normalized profiles of total heat flux in a fuel cell stack for open loop and closed loop, (a) Over 5,500 s of test period, (b) Over the first 200 s after fuel composition transition

410 The normalized profiles of the total heat flux in the fuel stack for both operation modes are shown in 411 Figure 13. The initial values for the open loop and closed loop tests were 62 kW and 61 kW respectively. As 412 shown in Figure 13a, the crossover point in the heat flux was achieved approximately at 1,300 s after using 413 humidified methane, as the heat flux dropped to about 50% from their initial values. Owing to the lower cathode 414 inlet air temperature at higher airflow within the first 1,300 s, the total heat flux in the open loop case was higher 415 than in the closed loop. These trends are clearly represented by the dynamic response over the first 200 s as shown 416 in Figure 13b, indicating a 17% reduction in the open loop system and near to 30% reduction in the closed loop 417 system. However, a 12% reduction in cathode air mass flow over 5,500 s of the open loop test ultimately resulted 418 in an 80% decrease in the total heat flux. In contrast, with constant cathode air flow, the total heat flux for the 419 closed loop only reduced by about 60% from the initial values.

427

#### 421 Fuel Cell Heat Generation

422 The detailed response in heat generation (HG) on an area specific basis is shown in Figure 14. In this 423 work, heat generation for an operating fuel cell was calculated using Eq. 5. 424

(5)

425 Fuel cell heat generation:

426  $HG = HG_{cell} + HG_{SMR} + HG_{WGS}$ 

428 The terms of  $HG_{cell}$ ,  $HG_{SMR}$ , and  $HG_{WGS}$  respectively represent the by-product heat generation from the electrochemical oxidation polarizations, heat generation from steam methane reforming (which is negative), and 429 430 water-gas shift reactions.

431 As illustrated in Figure 14a, heat generation at the beginning of the cell reduced dramatically immediately 432 after the fuel composition switch, following by a slow increase toward the new steady state at 5,500 s. The heat 433 generation profile obtained from the open loop was similar to the closed loop profiles, as shown in Figure 14b. 434



435 Figure 14: Fuel cell heat generation as a function of time, (a) Open loop transient response (b) Comparison of 436 open loop and closed loop transients

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438 The magnitude of heat generation at the initial steady state in Figure 14b matched the magnitude of heat 439 flux shown in Figure 12b. Therefore, the system exhibited fairly constant solid and gas temperature performance, 440 respectively presented in Figures 5 and 8. However, as the fuel was changed to humidified methane, heat 441 generation at the cell entrance reduced significantly from 0.7 W/cm<sup>2</sup> to about -1.4 W/cm<sup>2</sup>. This initial transient 442 was promoted by heat utilization in the steam methane reforming reaction. As compared to the heat flux profile 443 in Figure 12b, no significant fluctuation in heat flux was observed at 1 s after the step change due to delay in 444 temperature dynamics.

445 Heat generation at the cell entrance shown in Figure 14b increased gradually as the test progressed. To 446 be specific, the magnitude of heat generation at the beginning of the cell decreased with decreasing methane 447 conversion as the fuel cell solid temperature decreased. Because of the insignificant difference in CH<sub>4</sub> mole 448 fraction at 1 s between the open loop and closed loop tests (Figure 15), the same profiles of heat generation density 449 were obtained. However, heat generation at the beginning of the cell from both operations started to deviate slowly 450 throughout the test, which reflected to the trends in CH<sub>4</sub> mole fraction and the corresponding fuel cell solid 451 temperature.

The same impacts were also demonstrated at around center of the cell length. Heat generation decreased 452 453 over time as more CH<sub>4</sub> mole fraction shifted toward the elevated temperature region at the second half of the fuel 454 cell length. In contrast, heat generation at the back of the cell gradually increased over the time studied due to 455 electrochemical reactions. This behavior was found to be consistent with the trends in current density.

456 Since the inlet temperature at the end of the open loop test was higher than in the closed loop, more methane reforming was facilitated. Hence, the heat generation in the open loop case was more strongly negative, 457 458 as depicted by the profile at 5,500 s in Figure 14b. At 5,500 s, the total average heat generation of the stack was 459 14.5 kW, which was comparable to the 14.7 kW of heat flux shown in Figure 12, indicating that the system achieved a new steady state. 460

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#### 464 3.3. Transient Analysis of Fuel Compositions, Current Density, and Fuel Utilization

#### 466 *Methane*, $CH_4$

467 The variations in methane mole fraction along the length of the cell are presented in Figure 15. As shown 468 in Figure 15a, methane was consumed in the system over the entire course of the experiment, reducing from the 469 inlet composition at node 1 to near zero mole fractions at the cell outlet. Meanwhile, Figure 15b compares the 470 changes in  $CH_4$  mole fraction resulting from the open loop and closed loop operations. 471



Figure 15: Methane mole fraction as a function of time, (a) Open loop transient response (b) Comparison of open loop and closed loop transients

Within the first 2,450 s after the step change in fuel composition,  $CH_4$  reduced more slowly in the variable turbine speed operation, as compared to the constant turbine speed operation. The lower  $CH_4$  consumption (in open loop) was mainly due to lower fuel cell solid temperature as demonstrated in Figure 5a. The increased cathode air mass flow during the first transient period in the open loop reduced solid temperature. As such, methane reforming decelerated, resulting in lower  $CH_4$  consumption. In contrast, faster conversion of methane in the closed loop test was promoted by its higher operating temperature, as shown in Figure 5.

The reduction in cathode air mass flow in the open loop system subsequently induced the rapid depletion of CH<sub>4</sub> in the fuel cell subsystem. As shown in Figure 15b, the open loop dynamics obtained at 2,450 s followed the closed loop dynamics very closely. Both methane mole fraction and solid temperature crossover points were identical, suggesting close coupling of reforming to solid temperature. If this is the case, composition transients could be mitigated using cathode airflow modulation, which has been shown to have a strong impact on solid temperature.

#### 488 Other Fuel Composition Gradients, Current Density, and Fuel Utilization

489 The corresponding effects on other fuel constituents, such as CO,  $H_2$ , and  $H_2O$  are respectively presented 490 in Figure 16 through Figure 18. The slower and consequent rate limiting CH<sub>4</sub> reforming influenced the formation 491 and consumption of the other fuel components in water-gas shift and electrochemical oxidation.

492 Because of higher temperature at the initial steady state shown in Figure 5, the system was more 493 dominated by endothermic methane reforming rather than water-gas shifting. Therefore, both CO and H<sub>2</sub> mole 494 fraction at the first half of the cell length shown in Figures 16 and 17 increased with decreasing CH<sub>4</sub> mole fraction 495 (Figure 15). As expected, H<sub>2</sub>O mole fraction shown in Figure 18, significantly reduced in the beginning of the 496 cell. Due to less CH<sub>4</sub> in the downstream region, CO and H<sub>2</sub> mole fraction reduced gradually when a transition in 497 the water-gas shift reaction toward the product side and electrochemical oxidation of H<sub>2</sub> became more significant. 498 Higher formation of H<sub>2</sub>O that was reflected by consumption of H<sub>2</sub> was still observed at the end of the cell.

499 Immediately after the fuel composition change, the current density shown in Figure 19 increased to a 500 maximum value around node 6 to node 8. The  $H_2$  mole fraction was also at the maximum (Figure 17), suggesting 501 the formation of H<sub>2</sub> from steam methane reforming was significantly higher than its consumption in the 502 electrochemical oxidation. High CO generation rate provides further evidence of sufficient reforming to maintain 503 the water gas shift equilibrium toward the reactant side (Equation 2). Despite the maximum localized current 504 density, the fuel utilization was at a local minimum near node 6, as shown in Figure 20 for the 1 s curve. This 505 clearly indicated that large amounts of thermal energy in the fuel cell were being converted to chemical energy in 506 this region. Hence, this caused more reduction in solid temperature at the beginning of the cell. As there was less 507 energy in the cell, represented by lower temperature and thereby lower sensible heat, methane became more 508 distributed and the H<sub>2</sub> partial pressure reduced.

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Figure 16: Carbon monoxide mole fraction as a function of time, (a) Open loop transient response (b) Comparison of open loop and closed loop transients



Figure 17: Hydrogen mole fraction as a function of time, (a) Open loop transient response (b) Comparison of open loop and closed loop transients

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Figure 18: Water mole fraction as a function of time, (a) Open loop transient response (b) Comparison of open
 loop and closed loop transients

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The open loop system experienced a solid temperature drop by 50 K drop in just 250 s. Following the deceleration of  $CH_4$  reforming at more rapid solid temperature drop in the open loop case, the reduction in CO and  $H_2$  mole fractions were seen to be more apparent in the open loop test as compared to the closed loop test. The corresponding conductivity likewise reduced. The maximum current density region significantly shifted from the beginning of the cell, before the fuel composition switch, to further down the cell towards the end of the test. The same current density profiles were observed in the open loop and closed loop tests within the first 250 s due to very slight differences in the solid temperature and fuel composition dynamics.

525 As approaching a new steady state in the open loop case approximately at 5,500 s, the CO and  $H_2$  mole 526 fractions in the open loop case did not reduce as much as in the closed loop. The open loop test demonstrated 527 higher maximum CO and  $H_2$  mole fraction at 5,500 s, respectively with 22% and 12% relative differences to the 528 maximum value in the closed loop case. This was attributed to the higher solid temperature in the open loop case. 529 The higher temperature would favor reforming and inhibit water gas shift as written in Equation 2, both of which 530 result in higher CO partial pressures.

In fact, the maximum current density at 1,500 s and 5,500 s were also nearly identical and the curves were close, having come to equilibrium much faster than in the closed loop case. In contrast, an 8% higher current density was still noticeable at 5,500 s in the closed loop test, as a consequence of continuous changes in temperature and fuel composition partial pressures. At 5,500 s, current density increased with reduction in  $H_2$ mole fraction near the cell exit, both of which promoted higher fuel utilization.



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Figure 19: Fuel cell current density as a function of time (a) Open loop transient response (b) Comparison of open loop and closed loop transients



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#### 545 *3.4. Transient Analysis of Fuel Cell Thermal Energy Storage and Fuel Cell Thermal Effluent* 546

547 The dissipation of fuel cell stored thermal energy and the corresponding fuel cell thermal effluent in 548 response to the fuel composition changes are shown in Figure 21, where dissipation represents the change in rate 549 of thermal energy being stored in the fuel cell stack. The dissipation term was calculated based on the rate of 550 change in fuel cell average temperature multiplied with the total stack heat capacity as function of temperature (2625 kJ/K at the initial condition), as summarized in Eq. 6. The initial fuel cell stored thermal energy shown in 551 552 Figure 21a reduced immediately as a result of the accelerated steam methane reforming. The increasing  $H_2$ 553 generation at 1 s clearly illustrated the associated dynamic conversion of thermal energy to chemical energy, 554 which subsequently resulted in lower fuel utilization within increased current density region. Such transients 555 finally caused a dramatic increase in fuel cell thermal effluent exiting the system indicated in Figure 21b. The 556 system experienced a significant reduction in solid temperature as the stored thermal energy decreased. As compared to the closed loop transients, the dissipation of stored thermal energy in the open loop case recovered 557 more gradually before the crossover point at 1,300 s. However, the new steady state in the open loop was achieved 558 559 far quicker than in the closed loop due to the coupling of the cathode air mass flow in a longer test run.

The difference in the fuel cell thermal effluent between the open loop and closed loop cases shown in Figure 21b was attributed primarily to the difference in dissipation of stored thermal energy. The dynamic fuel 562 cell thermal effluent remained higher than the initial value for about 1,600 s before decreasing to the new steady 563 state, following non-minimum phase behavior (transient inverse response) as opposed to its initial steady state 564 performance [17]. This ultimately caused the initial rapid increase in turbine speed and cathode air mass flow 565 shown in Figure 3. The overshoot response in fuel cell thermal effluent was identified as one of the main 566 operational challenges to fuel flexibility in gas turbine hybrid systems. It is critical to develop novel control 567 strategies to limit such overshoot and expand the range of fuel composition changes that could be implemented. 568 Nonlinear control models are required to describe the entire trajectories of the fuel cell thermal effluent.

- 570 Dissipation of fuel cell stored thermal energy:
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Figure 21: Changes in fuel cell stored energy and fuel cell thermal effluent for the open loop and the closed loop
 tests

#### 578 4. Conclusions

579 The detailed dynamic characterization of an SOFC/GT hybrid system under a transition from syngas to 580 humidified methane revealed:

- a. An extensive conversion of fuel cell stored thermal energy immediately after the step change, resulting in a sudden transient inverse response in fuel cell transferred heat,
- b. A spike in thermal effluent from the fuel cell post combustor system to the turbine, which occurred in spite of a dramatic reduction in both heat generation and heat flux, which was dominated by the conversion of stored thermal energy to chemical energy,
- c. A substantial solid temperature drop (50 K) in the fuel cell 250 s after the step change due to propagation
   impacts of increased cathode air mass flow,
  - d. A maximum of 17 K/cm solid temperature gradient in the fuel cell from an initial state of 12 K/cm,
- 589 e. Significant differences in the distributed performance of temperature, heat flux, heat generation,
   590 composition gradients, fuel utilization, and especially current density,
- f. Substantial cathode air flow coupling effects over the course of the entire study from the initial change to the new steady state,
  - g. Close coupling between the steam methane reforming and solid temperature.

The open loop operation demonstrated significant variations in turbine speed and cathode mass flow. As much as 6% turbine speed variation from nominal condition was observed. Hence, the system experienced a 12% cathode air mass flow change. Therefore, fuel cell temperature, heat flux, and temperature gradient across the fuel cell length changed accordingly due to highly coupling effects. However, this perturbation helped the system to achieve a new steady state faster than the closed loop system.

As compared to the closed loop, operating the SOFC/GT hybrid system with turbine speed changes resulted in less detrimental performance after the crossover points, with higher temperature, lower current density, and lower fuel utilization. We also found that small changes in turbine efficiency at higher speed in the open loop over the closed loop tests affected fuel cell heat flux through variations in cathode air inlet temperature. Overall, all

key fuel cell parameters were highly coupled and solid temperature appeared to be the primary linking event ofmany mechanisms.

606 In conclusion, fuel cell solid temperature control is critical to avoid excessive temperature gradients, which 607 could not be tolerated by the fuel cell material. Severe cooling effects might be also localized at the beginning of the cell, depending on the initial methane content. So, a shorter cell lifetime would be expected due to increasing 608 degradation risk since the system was operated at a lower temperature. Therefore, future dynamic control systems 609 610 for SOFC/GT hybrid systems must include effective thermal management, most likely implemented through cathode air flow management. Nonlinear control strategies must be employed to deal with the system complexity 611 adequately. It is clear from the comparison of open and closed loop transient study that cathode airflow has the 612 613 strongest linking effect on fuel composition changes, and it holds the greatest promise for active control in fuel

- 614 flexible systems.
- 615

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