Advanced Process Control and Dynamic Optimization of Reversible Solid Oxide Cell Systems for Performance and Long-Term Health

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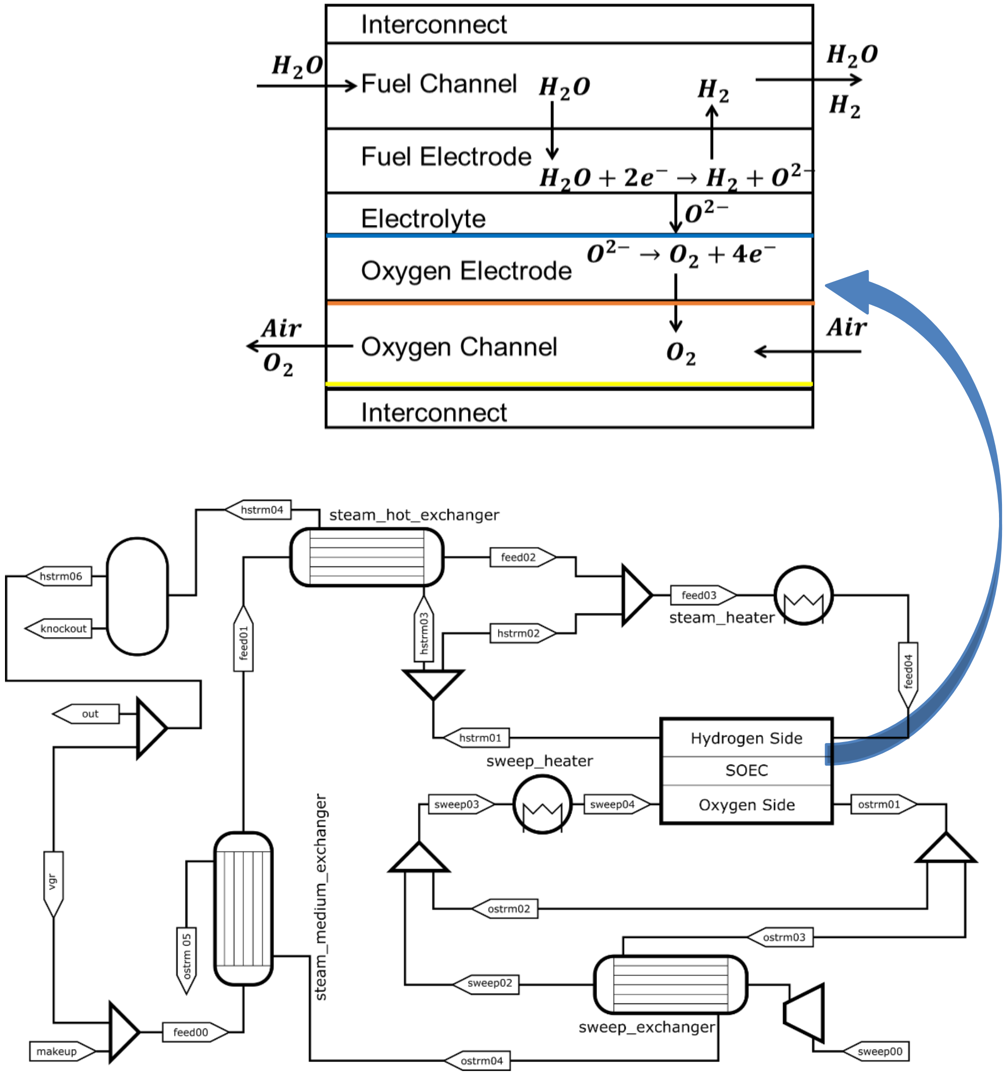
SOC-based System for Hydrogen and Power Production

- **Key Challenges**
  - Solid Oxide Cell (SOC)-based systems for hydrogen and power production **need to operate flexibly** with fluctuations in market electricity prices.
  - How can we **best operate and control** SOC-based systems for **mode-switching (H_2/power)**, while **minimizing degradation** over long-term flexible operation?

- **Technical Approach**
  - Develop a **first-principles dynamic model** of SOC-based systems
  - Develop **classical and advanced process controls** for effective thermal management and mode-switching operation
  - Develop first-principles **degradation** sub-models to **quantify the impact of flexible operation** on cell health
  - Optimize **long-term performance** and **health** of flexible SOC-based systems

Dynamic Model of SOC-based System for Mode-Switching

- **IDAES** open-source, equation-oriented modeling and optimization framework (Lee et al., 2021)
- **SOC dynamic model** (Bhattacharyya et al., 2007)
  - First-principles, non-isothermal, planar cell
  - 2D electrodes, electrolyte, and interconnect
  - 1D fuel and oxygen channels
  - Operates in fuel cell and electrolysis modes
- **Dynamic SOC-based system model** (Allan et al., 2023)
  - **H_2 fueled** in fuel cell mode
  - **Vent gas recirculation** with purge
  - **Condenser** to remove water from H_2-side off-gas
  - Equipment models for **thermal management**
    - 1D multi-pass crossflow recuperative heat exchangers
    - 1D crossflow trim heaters
  - System **performance constraints**



Block flow diagram of H_2–fueled SOC-based IES for Mode-Switching Operation

- Lee, A., et al.., J Adv Manuf Process 2021, 3(3) (2021).
- Bhattacharyya et al., Chem Eng Sci, 62, 4250-4267 (2007).
- Allan, D.A., et al., In Proc. FOCAPO/CPC (2023).

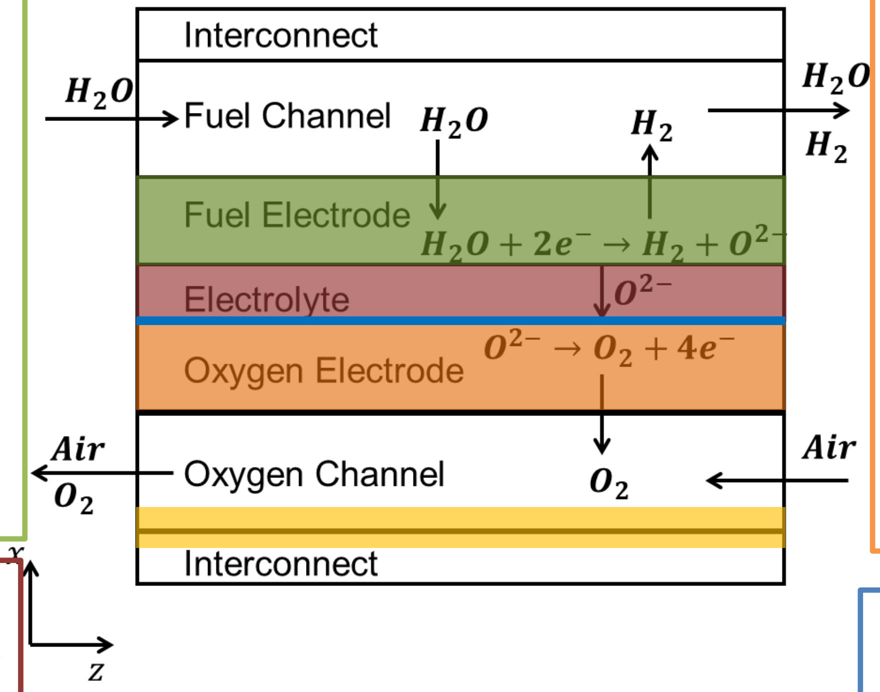
SOEC Microstructure Chemical Degradation Modeling

Fuel electrode nickel (Ni) agglomeration

- Ni particles grow with time under high temperature operation
- Ni_2OH formation drives the process
- Surface-diffusion – Ostwald ripening

$$\frac{d(d_{Ni})}{dt} = C\frac{X_{Ni}}{X_{YSZ}A_{YSZ}d_{Ni}^{6}}\left(\frac{Y_{H_2O}}{Y_{H_2}^{0.5}}\right)\exp\left(-\frac{E_a}{RT}\right)$$

Refs: J. Sehested et al. / *Applied Catalysis A*: General 309 (2006) 237–246



Lanthanum zirconate (LZO) scale growth

- At oxygen electrode under oxidizing conditions and high temperatures driven by high P_{O_2}

$$LaMnO_3 + ZrO_2 + 0.25O_2 \rightleftharpoons 0.5La_2Zr_2O_7 + MnO_2$$

- Parabolic growth law

$$\frac{dl_{LZO}(t)}{dt} = \frac{K_{g,LZO}}{2l_{LZO}(t)X_{0,LZO}\rho_{LZO}}exp\left(\frac{E_{LZO}}{RT}\right)$$

Refs: A. Kamkeng, and M. Wang. / *Chemical Engineering Journal* 429 (2022): 132158

YSZ electrolyte phase transformation

- Phase transformation of YSZ from cubic to tetragonal structure
- Results in decrease in electrolyte conductivity

$$\sigma_{El} = \sigma_{El,0}\left[\lambda + (1-\lambda)\exp\left(-\frac{t}{\tau}\right)\right]$$

Refs: Jiang et al. *Journal of the American Ceramic Society* 82(11):3057 - 3064

Chromium oxide scale growth

- Oxidation of chromium interconnect-oxygen electrode boundary
- Parabolic growth law

$$\frac{dl_{COS}(t)}{dt} = \frac{K_{g,COS}}{2l_{COS}(t)X_{0,COS}\rho_{COS}}exp\left(\frac{E_{COS}}{RT}\right)$$

Refs: D. Larrain et al. / *Journal of Power Sources* 161 (2006) 392–403

LSM-YSZ phase coarsening

- Driven by Mn^{2+} diffusion from LSM surface toward LSM-YSZ interface
- Results in loss of TPB length
- Model derived by assuming Fick's law diffusion of Mn^{2+}

$$\frac{L_{TPB}}{L_{TPB,0}} = 1 - 2 \times \left(\frac{t \times D_{LSM}}{\pi}\right)^{1/2}$$

Refs: A. Kamkeng, and M. Wang. / *Chemical Engineering Journal* 429 (2022): 132158

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Impact of Chemical Degradation on Cell Temperatures

Chemical degradation depends on operating strategy

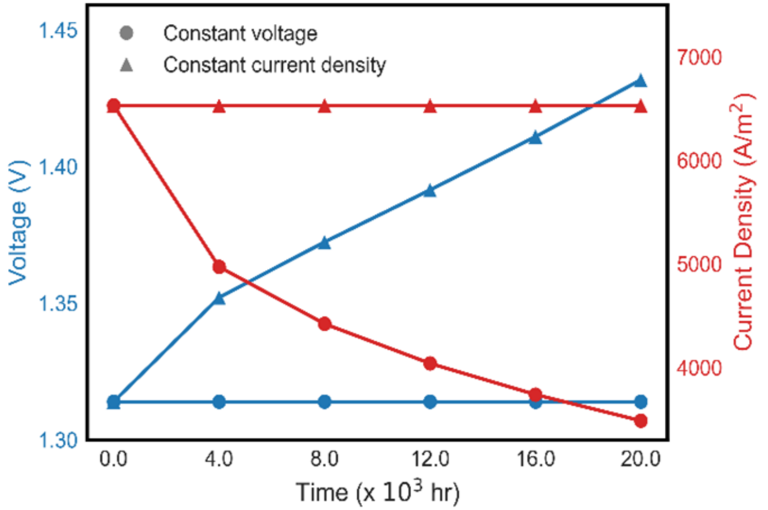
- **Constant voltage (potentiostatic)**
- **Constant current density (galvanostatic)**

What happens if the initially optimal cell operating conditions are held constant?

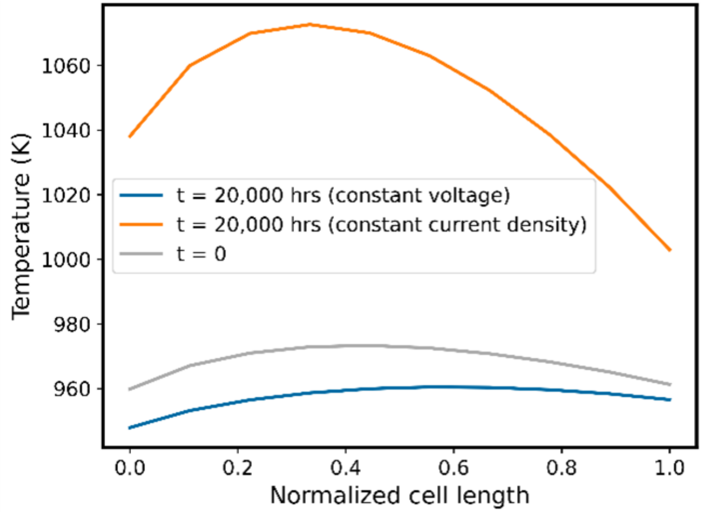
- Initial steady state optimization profiles held constant for 20,000 hours
- **Inlet temperatures held constant**

Impact on cell performance

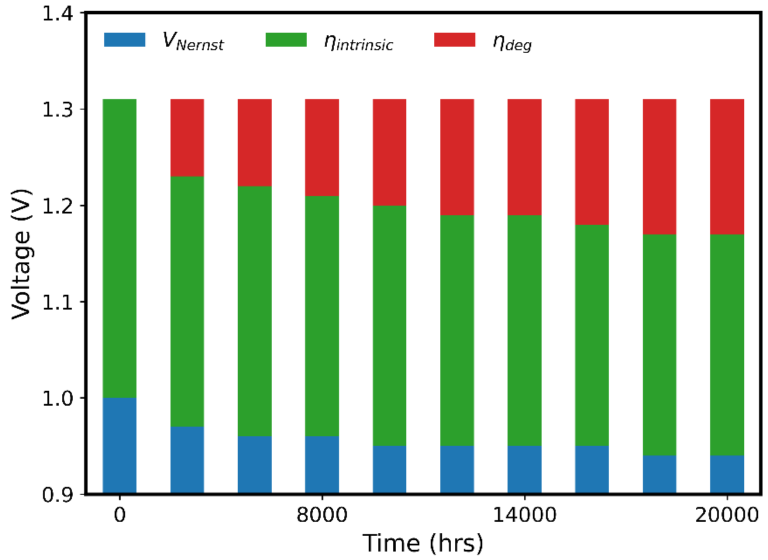
- Potentiostatic operation results in 42% decrease in current density
- Galvanostatic operation results in 8.3% increase in cell terminal voltage



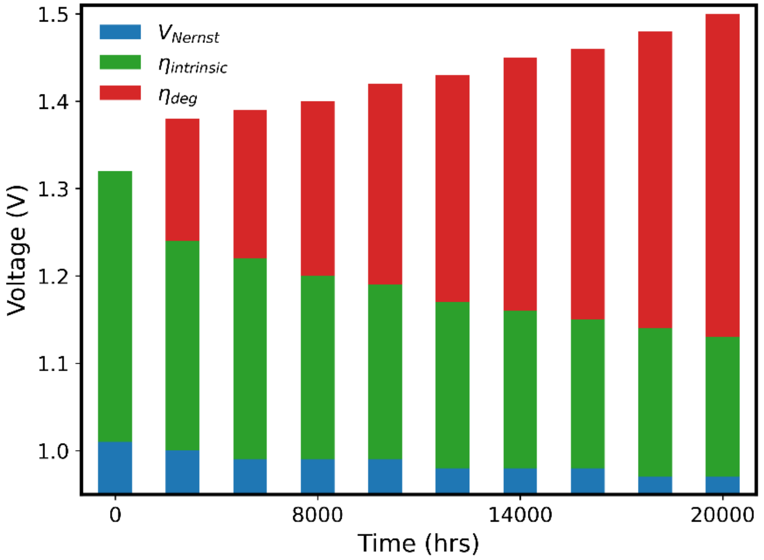
Operating profile of current density and voltage under potentiostatic and galvanostatic operation



Temperature profile under potentiostatic and galvanostatic operation after 20,000 hr of operation



Voltage losses under potentiostatic operation



Voltage losses under galvanostatic operation

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Optimizing Long-Term SOEC System Operation

Case 1: Maximize Integral Efficiency

$$max \frac{1}{t_f - t_0} \int_{t_0}^{t_f} \eta_t dt$$

$st.$

$$h(x) = 0$$

$$\frac{dR}{dt} = f_R(x,t)$$

$$\eta_t = \frac{HHV(\dot{m}_{H_2,t})}{P_{in,total,t}} \qquad \forall t$$

Case 2: Minimize Final Degradation

$$min\ \Delta V\left(\theta_{t_f}\right)$$

$st.$

$$h(x) = 0$$

$$\frac{dR}{dt} = f_R(x,t)$$

$$\theta_{tf} = \theta_{\ t0} + \int_{t_0}^{t_f} \dot{\theta}(x,\theta)dt$$

Decision variables at each time point:

1. Feed heater duties
2. Sweep heater duties
3. Sweep blower flowrate
4. Feed exchanger flowrate
5. Feed recycle ratio
6. Sweep recycle ratio

Case 3: Minimize Levelized Cost of Hydrogen (LCOH)

$$LCOH = \frac{CRF_{BOP}CC_{BOP} + \sum_{i=1}^{R} CRF_{stack,i}CC_{stack} + OC + EC}{m_{H_2,\text{lifetime}}}$$

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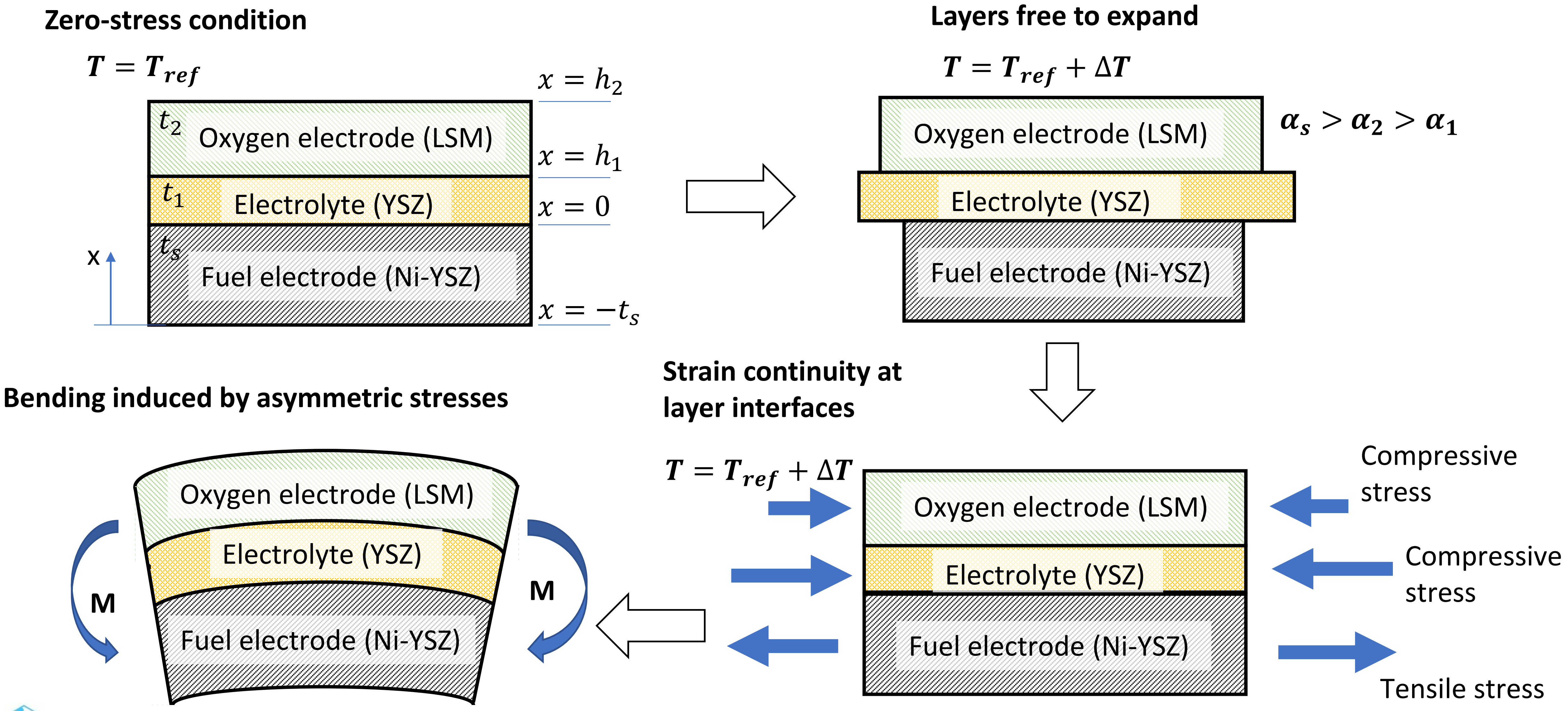
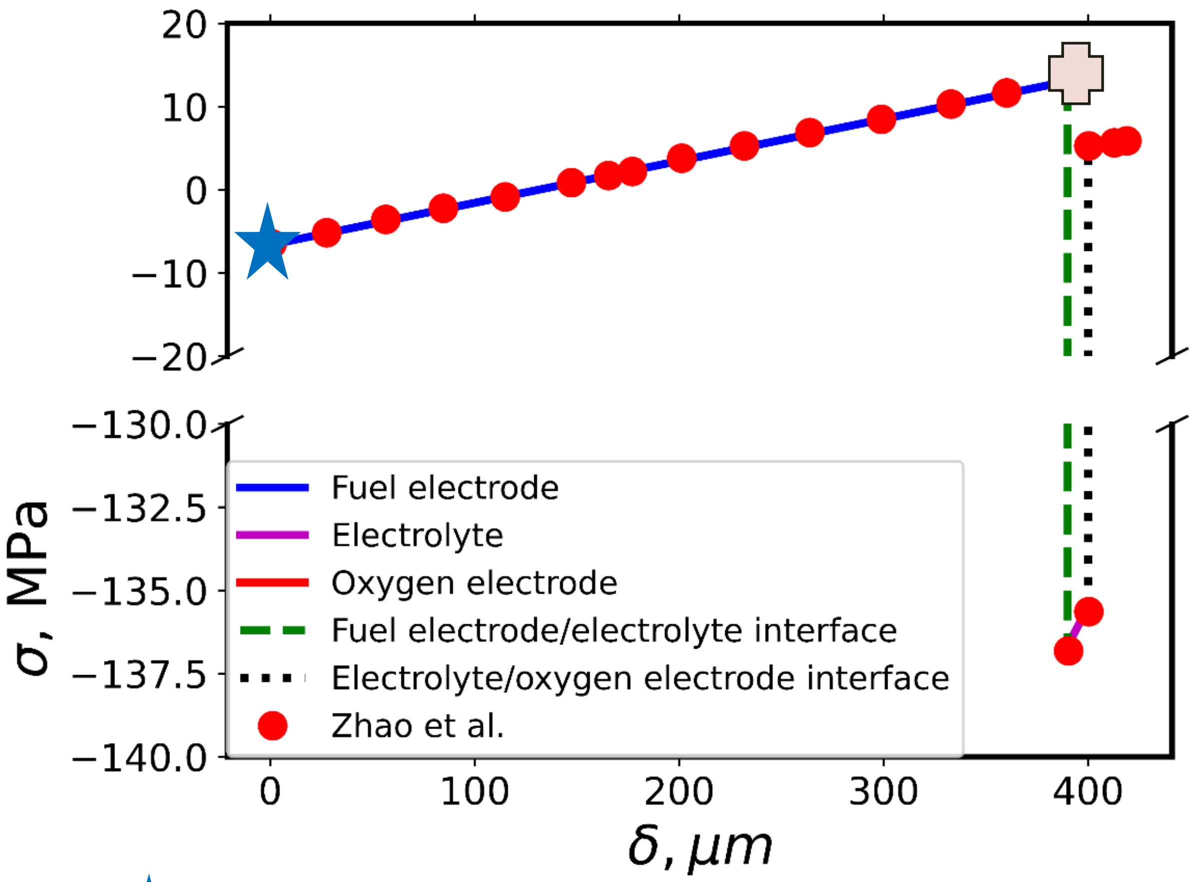
Quasi-Steady State Optimization Results for 3 Objective Functions under Galvanostatic, Potentiostatic, and Flexible Operation for Low- and High-Price Electricity Markets

		Electricity Price = 0.03 $/kWh		Electricity Price = 0.3 $/kWh	
Operating Profile	**Objective Function**	**Replacement Schedule (years)**	**LCOH ($/kg H_2)**	**Replacement Schedule (years)**	**LCOH ($/kg H_2)**
Galvanostatic Operation	Minimize terminal degradation	5	2.00	5	13.00
	Maximize Integral Efficiency	2	2.29	2	11.92
	Minimize LCOH	5	1.93	2.5	11.84
Potentiostatic Operation	Minimize terminal degradation	3	2.11	3	12.51
	Maximize Integral Efficiency	2	2.30	2.0	11.93
	Minimize LCOH	3	2.05	2.0	11.91
Free Operation	Minimize terminal degradation	5	1.99	5	13.01
	Maximize Integral Efficiency	3	2.02	2.5	11.78
	Minimize LCOH	5	1.92	2.5	11.78

Physical Degradation: Thermal Stress Evolution



Understanding Thermal Stress Distribution



★ Compressive stress at fuel electrode / Channel interface

✚ Tensile stress (max) at fuel electrode / Electrolyte interface

- Sustained operation at high temperature affects thermal and mechanical properties causing localized **stress build-up**.
- Ceramics are more resilient to compressive stresses than tensile stresses.
- Steep changes in stress profiles and directionality are observed around interfaces.

Maximizing Efficiency under Stress Constraint

$$max \frac{1}{t_f - t_0} \int_{t_0}^{t_f} \eta_t dt$$

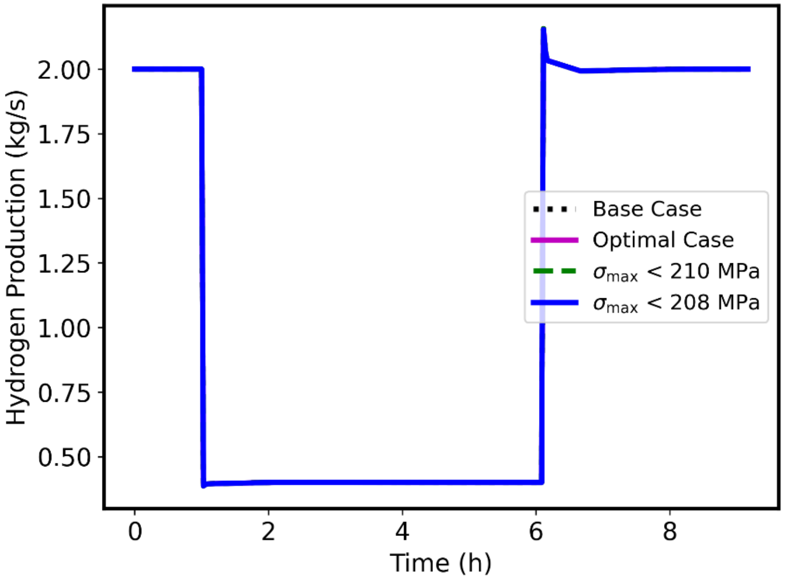
$st.$

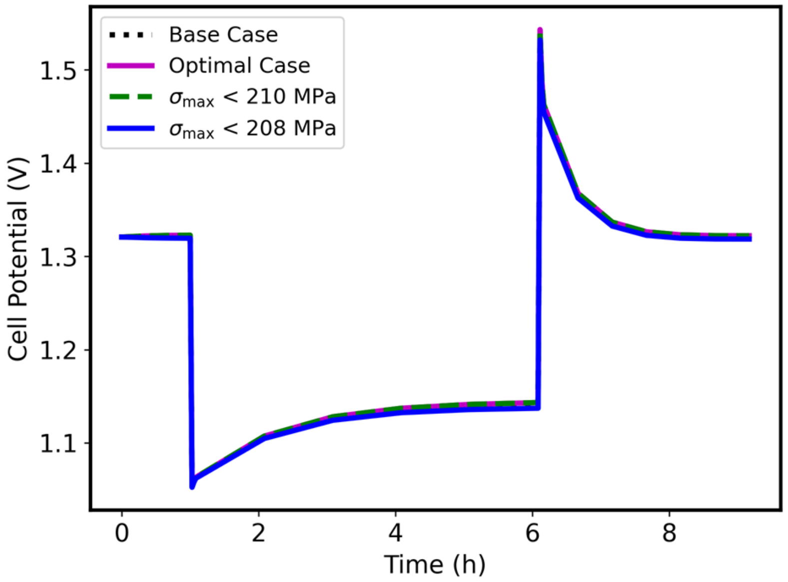
$$h(x) = 0$$

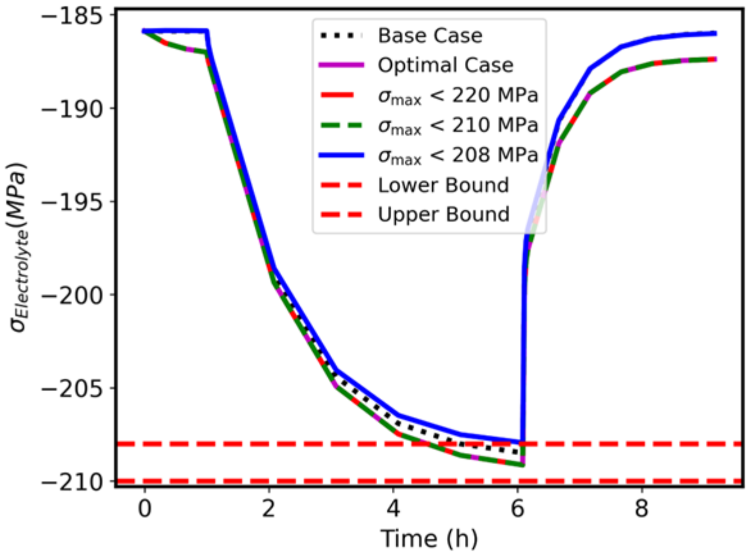
$$\frac{dR}{dt} = f_R(x,t)$$

$$\sigma^i \leq \sigma^i_{max}[MPa]$$

$$\eta_t = \frac{HHV(\dot{m}_{H_2,t})}{P_{in,total,t}} \qquad \forall t$$







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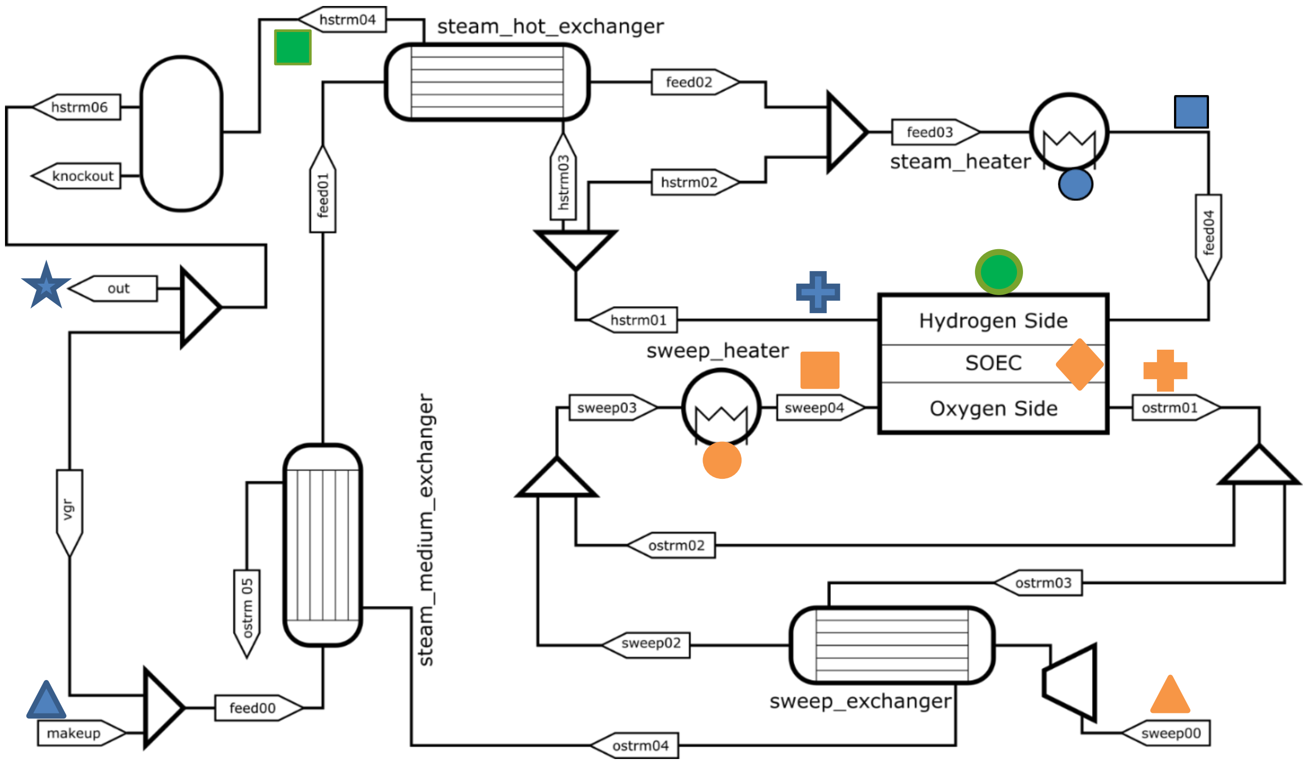
Process Control for SOC-based System Mode-Switching

- **Classical Control: Proportional-Integral-Derivative (PID)**
- **Nonlinear Model Predictive Control (NMPC)**
  - Well-suited to highly interactive manipulated variables and constraint handling



Controller	Manipulated Variables (MVs)	Controlled Variables (CVs)
PID, NMPC	Cell potential	Outlet Water Concentration
PID, NMPC	Steam/H_2 feed rate	H_2 production rate
PID, NMPC	Feed heater duty	Feed heater outlet temperature
PID, NMPC	Sweep heater duty	Sweep heater outlet temperature
PID, NMPC	Steam heater outlet temperature setpoint*	SOC steam outlet temperature
PID, NMPC	Sweep heater outlet temperature setpoint*	SOC sweep outlet temperature
PID, NMPC	Sweep feed rate	SOC temperature
NMPC	Feed recycle ratio	
NMPC	Sweep recycle ratio	
NMPC	Vent gas recirculation (VGR) recycle ratio	
NMPC	H_2/H_2O ratio in make-up	

**artificial control variables*

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- Allan, D.A., et al., In Proc. FOCAPO/CPC (2023).
- Dabadghao, V., Ph.D. Thesis, CMU (2023).

Nonlinear Model Predictive Control (NMPC)

- **NMPC Objective Function**
  - Developed for setpoint transition
  - Penalties for cell temperature curvatures related to degradation

$$f_{obj} = \sum_{i=0}^{N} \rho_{H_2} \left(y_i - y_i^R \right)^2 + \sum_{i=0}^{N} \sum_{j \in J} \rho_j \left(u_{ij} - u_{ij}^R \right)^2 + \sum_{i=0}^{N} \sum_{k \in K} \rho_k \left(x_{ik} - x_{ik}^R \right)^2 + \sum_{i=1}^{N} \rho' \left(v_i - v_{i-1} \right)^2 + \sum_{i=0}^{N} \sum_{z=1}^{z_L} \rho_M \left(\frac{\partial^2 T_{iz}}{\partial z \partial t} \right)^2$$

Trajectory tracking of H_2 production rate

Deviations of manipulated (u_{ij}) and controlled variables (x_{ik}) from reference values

Rate of change penalties on trim heater duties

Penalties for cell temperature curvatures

- **Solution Approach**
  - Classical Control: PETSc variable step implicit Euler DAE solver
  - NMPC: **Full-discretization** NLP with IPOPT solver
- **NMPC Problem Size**
  - Approximately 16,000 equations and variables
  - Average solution time of 35.5s for a prediction horizon of 750s
  - Horizon size N=5 with a sampling time of 150s

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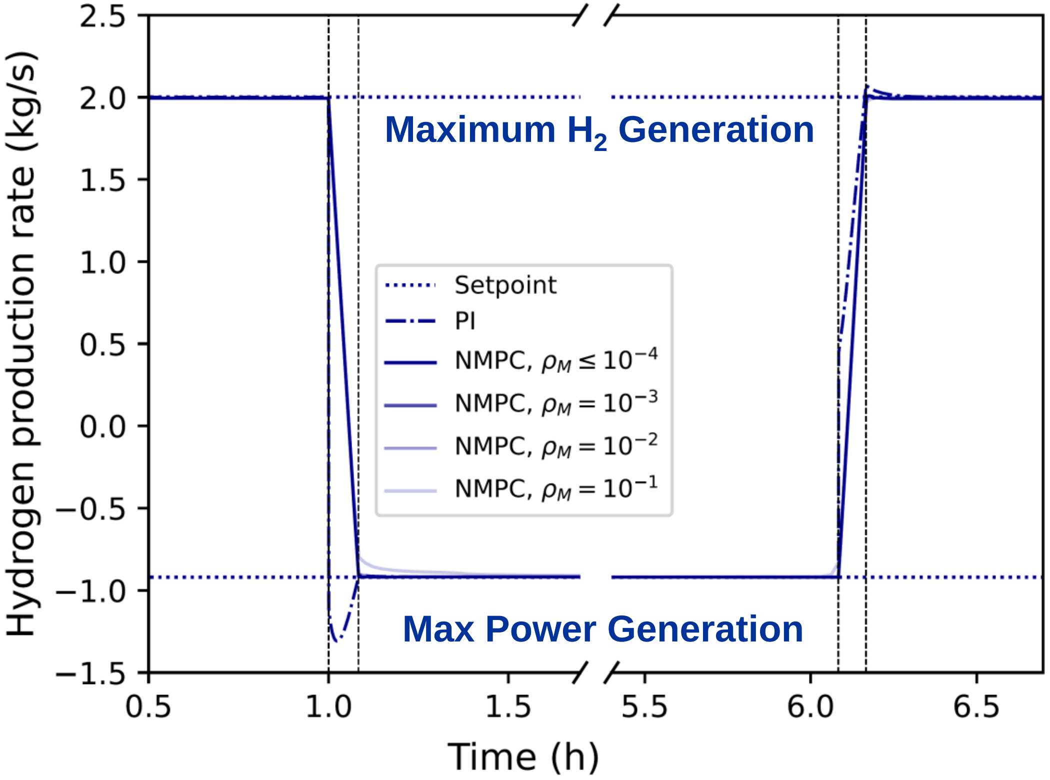
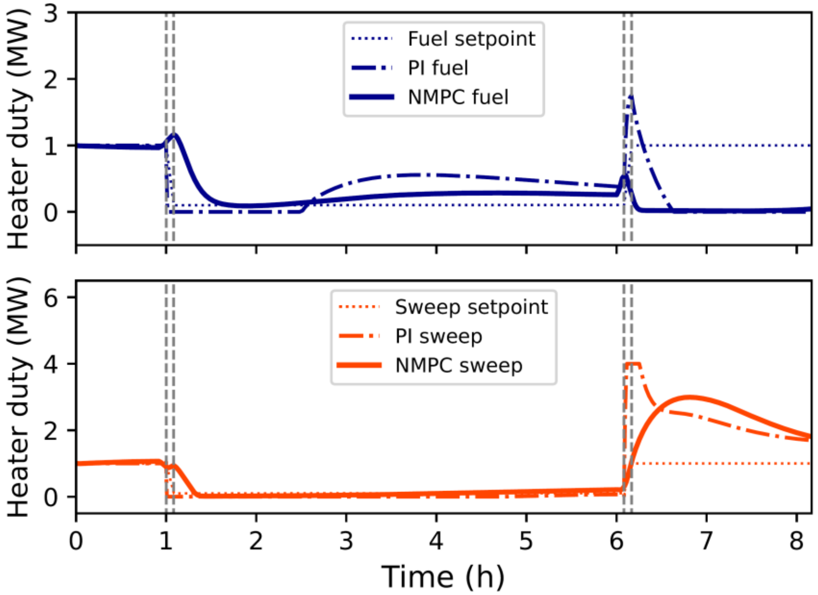
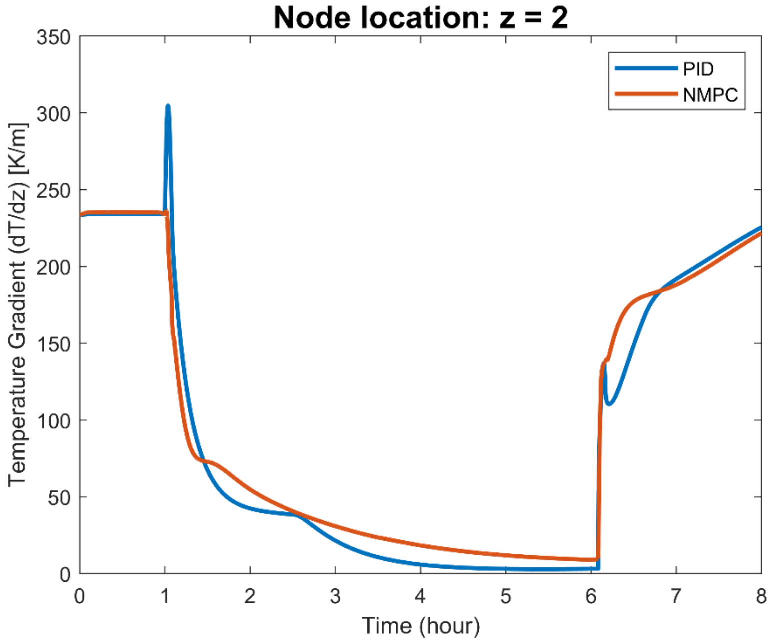
Dynamic Simulation and Control Results for Ramping Operation







- **Classical PI control** of H_2 production rate shows **overshoot**, not exhibited by NMPC
- **NMPC** yields **smoother heater duty profiles** than PI control
- **NMPC** yields **smoother SOC temperature gradient** and lower spatial extremum magnitude than PI control

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Conclusions

- **Long-term SOEC optimization** considering **chemical degradation** can be used to optimize **stack replacement schedule** and **operating trajectory.**
  - In **high-price** electricity markets, SOEC system **efficiency is maximized** at the expense of **more frequent stack replacements.**
  - In **low-price** electricity markets, SOEC system is operated under **more conservative conditions** to **reduce the frequency of stack replacements.**
- **Dynamic optimization** considering **physical degradation** can be used to optimize SOEC operating trajectories to **satisfy spatio-temporal stress constraints** without much sacrifice in the overall efficiency.
- **NMPC** can explicitly **restrict temperature gradients/curvatures or other constraints** compared to classical control.
- **IDAES** is a powerful modeling and optimization tool for analyzing **how to best operate and control** SOC-based systems for **mode-switching (H_2/power)**, while **minimizing degradation** over long-term flexible operation.

IDAES Team and Funding Acknowledgements

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github.com/IDAES/idaes-pse

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