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Self-adaptive heat management of solid oxide electrolyzer cell under fluctuating power supply



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ABSTRACT

Solid oxide electrolyzer cell (SOEC) can efficiently reduce CO_2/H_2O into CO/H_2 using renewable powers. But the fluctuating nature of renewable energy can significantly change the temperature field and cause thermal fatigue even mechanical failure. Herein, we propose a self-adaptive heat management method of SOEC by coupling SOEC with an in-situ thermochemical energy storage (TES) section. Co_3O_4/CoO redox pair is selected as the TES material for heat management during the exothermic and endothermic operating conditions. 2D dynamic models are developed to study the effects of TES section on the SOEC performances with a special attention on the temperature fluctuation. Results show that the addition of TES section can reduce temperature fluctuation by 73 %, from 119.2 K to 31.7 K. Besides, the maximum temperature gradient and the surface maximum temperature difference at 1.5 V are reduced to 2.62 K/mm and 30 K, respectively, which are 65 % and 82 % lower than those without TES section. An inlet temperature fluctuation of the cell, where the standard deviation is 8.51. This study provides a novel strategy for efficient heat management and safe operation of SOEC under fluctuating working conditions.

1. Introduction

To address the challenges of limited fossil fuel resources, climate change, and environment pollution, it is highly demanded to replace fossil fuels by renewable powers such as solar and wind energy [1,2]. However, renewable powers are intermittent, fluctuating, and site specific, limiting their wide and efficient application. Energy storage systems are thus needed to convert the fluctuating renewable powers into stable energy forms [3,4]. Solid oxide electrolyzer cell (SOEC) is one of the most competitive and prospective electrolysis technologies for converting H_2O-CO_2 mixture directly into syngas, which not only can be directly used as fuels, but also can be further synthesized into various hydrocarbons for the subsequent industry applications [5,6].

The high operating temperature (600–1000 °C) of SOEC can reduce the electrical energy requirement in electrolysis and increase electrochemical reaction activity [7]. But a high operating temperature also brings a big challenge to the heat management of SOEC especially when directly supplied with fluctuating renewable powers. The fluctuating nature of renewable energy can significantly change the temperature field of SOEC and cause thermal fatigue even mechanical failure [8,9]. Besides, a small temperature gradient should be maintained to avoid degradation acceleration and material failure. Therefore, advanced heat management methods are needed to inhibit temperature fluctuation of SOEC under dynamic operating conditions.

In recent years, various methods have been presented to regulate the temperature efficiently of reversible solid oxide cells and reduce the possibility of cell structure fracture caused by temperature gradient. In terms of structural design, Shi et al. [10] placed turbulators in the air channel to produce recirculation flow, which realized the enhancement of local convective heat transfer and decreased temperature gradient effectively especially that higher than 30 °C/cm. Rosner et al. [11] investigated the effect of SOFC design parameters on the thermal management, and found that increasing the rib width could effectively reduce thermal gradients. Yuan et al. [12] compared the effect of three different flow arrangements (co-, counter- and cross-flow) on cell performance and found that the cross flow had the most uniform

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Fig. 1. Working mechanism of co-electrolysis process coupling with TES.

temperature distribution. Kim et al. [13] proposed a novel interconnect design to the reduced thermal resistance. Based on 3-D thermo-fluid simulations, they found the temperature difference across the cell surface could be decreased by 35-60 °C at the repeating-unit scale. Perret [14] constructed independent channels to circulate water vapor in the electrolyzer cell stack, and achieved the purpose of absorbing heat from the electrolyzer cell or releasing heat to the electrolyzer cell by regulating the temperature of water vapor at the entrance. Dillig et al. [15] integrated high temperature heat pipes into SOEC stack to achieve temperature regulation in the stack using phase transition and migration of working medium in the heat pipe. In terms of reaction process control, Wang et al. [16] and Lin et al. [17] achieved absorbing excess heat by regulating ammonia cracking reaction and methane reforming reaction respectively, thus reducing the maximum temperature difference inside the stack. Li et al. [18] proposed a Gaussian process regression model to estimate the methane conversion rate, which avoided the problems of excessive temperature gradient and cell deformation and even fracture caused by too low methane conversion rate. Skafte et al. [19] applied alternating currents (a 90 % electrolysis and 10 % fuel cell cycle) on a SOEC, and found that this operating mode not only alleviated temperature change, but also Ni migration. In terms of material development, Shao et al. [20] proposed a novel thermal expansion offset approach for composite cathode material development by introducing negative thermal expansion materials into perovskite cathode materials. The newly developed cathode not only achieved excellent electrochemical performance, but also excellent thermomechanical stability as the thermal expansion coefficient (TEC) of the composite cathode was very close to that of the electrolyte, which greatly reduced the thermal stress and avoided cracks and mechanical failure of the cell. However, this strategy may involve a higher material cost and the long-term stability remains to be demonstrated.

Although above methods are effective in alleviating electrode fracture caused by temperature gradient and temperature change, the aircooling way is the most practical and economical alternative. Temperature fluctuation during dynamic operation of SOEC can be suppressed by controlling the air flow rate and temperature [21]. However, the ability to regulate SOEC temperature field by changing air flow rate is limited due to the small specific heat capacity of air. Wang et al. [8] investigated the influence of excess air ratio on temperature gradient and temperature variation and found that when the applied voltage was higher than 1.4 V, the excess air ratio must reach 25 to limit these two indicators within a reasonable range. Nevertheless, higher excess air ratio requires more pump power, which will reduce the system efficiency and increase the running cost. Aguiar et al. [22] reported that the maximum excess air ratio must be less than 14 to avoid significant additional energy costs. Cai et al. [23] found that the temperature controlled by air flow rate would lead to a large potential dynamic

response time constant, which was difficult to meet the demand of coupling with renewable energy. Therefore, it is urgent to develop an efficient temperature control method based on the heat source term to meet the dynamic change when using renewable energy as input.

When the SOEC operates below the thermoneutral voltage (TNV), the heat generation due to overpotential losses is lower than the heat demand for electrochemical reactions and SOEC will be in an endothermic state. When operating above the TNV, the heat generation is higher than the heat demand and SOEC will be in an exothermic state. Thermochemical energy storage (TES) material can absorb heat at high temperature through reduction reaction and release heat at low temperature through oxidation reaction, which was successfully used to protect the molten salt receiver in a concentrated solar plant [24]. When TES and SOEC are integrated, it can store the heat generated due to exothermic state of SOEC to compensate the heat demand for endothermic state of SOEC though oxidation reduction reactions, which will achieve adaptive temperature regulation. Besides, TES is maintaining solid-state in the reaction processes, which will not destroy the structure of SOEC. Mottaghizadeh et al. [25] integrated phase change material (PCM) and reversible solid oxide cell in series, but his idea was only limited to improving comprehensive energy utilization efficiency, and did not clarify the changes of physical parameters such as temperature and temperature gradient after the two are connected in parallel. Promsen et al. [26] coupled PCM into SOFC stack to improve the thermal conditions, and found that PCM could not only improve the temperature distribution but also suppress the temperature variation. However, this construction requires a separate channel for the PCM, which will increase the complexity of SOFC stack.

Despite some studies on the SOEC temperature control by employing thermal storage materials [25–29], the direct combination of TES with SOEC for temperature regulation under renewable energy input is still missing. In this paper, we propose a novelty heat management method by coupling SOEC with an in-situ TES section, which is placed into the air channel of a tubular SOEC model. We not only focus on the temperature regulation by TES, but also consider the fluctuating renewable power input. With a fluctuating PV power input, gas supply follows the electrical load to maintain a high gas utilization rate. We conduct detailed parametric studies to focus on analyzing the influence of the TES on the cell average temperature fluctuation reduction through a day and axial temperature gradient reduction.

2. Model description

A 2D-axisymmetric dynamic co-electrolysis SOEC model coupled with the TES is developed, including SOEC sub-model, TES sub-model and CFD sub-model. The CFD sub-model serves as a bridge to couple the heat produced or consumed by the SOEC sub-model with the heat

Table 1

Material parameters of SOEC.

Electronic conductivity $4.2 \times 10^7 exp(-1150/T)$ S/m Ni $4.2 \times 10^6 - 1065.3T$ S/m Ionic conductivity $5.2 \times 10^4 exp(-9681/T)$ S/m ScSZ $6.92 \times 10^4 exp(-9681/T)$ S/m YSZ $3.34 \times 10^4 exp(-10300/T)$ S/m Thickness (Ni + YSZ) m Cathode support layer 760 µm (Ni + YSZ) um (Ni + ScSZ) Cathode active layer 10 µm (ScSZ) um (ScSZ) Anode active layer 15 µm (LSM + ScSZ) um (ScSZ) Heat Conductivity Z.7 W/(m·K) Anode 9.6 W/(m·K) Anode 390 J/(kg·K) Electrolyte 300 J/(kg·K) Electrolyte 300 J/(kg·K) Density Um Scondo Cathode 6870 kg/m³ Electrolyte 2000 kg/m³	Parameters	Value or expression	Unit
LSM $4.2 \times 10^7 exp(-1150/T)$ S/m Ni $4.2 \times 10^6 - 1065.3T$ S/m Ionic conductivity $ScsZ$ $6.92 \times 10^4 exp(-9681/T)$ S/m ScSZ $6.92 \times 10^4 exp(-9681/T)$ S/m YSZ $3.34 \times 10^4 exp(-10300/T)$ S/m Thickness $Cathode support layer 760 µm (Ni + YSZ) µm (Ni + ScSZ) µm Cathode active layer 10 µm (ScSZ) Anode active layer 15 µm (LSM + ScSZ) µm (LSM + ScSZ) Heat Conductivity Z.7 W/(m.K) Electrolyte 2.7 W/(m.K) Heat Capacity W/(m.K) W/(m.K) Heat Capacity Z W/(m.K) Cathode 390 J/(kg.K) Electrolyte 300 J/(kg.K) Electrolyte 300 J/(kg.K) Electrolyte 2000 kg/m^3 Anode 6870 kg/m^3 $	Electronic conductivity		
Ni $4.2 \times 10^6 - 1065.3T$ S/m Ionic conductivity 5CSZ $6.92 \times 10^4 exp(-9681/T)$ S/m YSZ $3.34 \times 10^4 exp(-10300/T)$ S/m Thickness $(Ni + YSZ)$ ym Cathode support layer 760 μ m $(Ni + YSZ)$ μ m $(Ni + ScSZ)$ Electrolyte 10 μ m $(ScSZ)$ μ m $(ScSZ)$ Anode active layer 15 μ m $(LSM + ScSZ)$ μ m $(LSM + ScSZ)$ Heat Conductivity $Z.7$ $W/(m.K)$ Cathode 9.6 $W/(m.K)$ Anode 9.6 $M/(m.K)$ Heat Capacity $Z.7$ $W/(m.K)$ Cathode 390 $J/(kg.K)$ Electrolyte 300 $J/(kg.K)$ Density V V Cathode 6870 kg/m^3 Electrolyte 2000 kg/m^3	LSM	$4.2 \times 10^7 exp(-1150/T)$	S/m
$\begin{tabular}{ c c c } lonic conductivity $$CSZ$ 6.92 \times 104 exp(-9681/T) $$/m$ $$/m$ $$YSZ$ 3.34 \times 104 exp(-10300/T) $$/m$ $$/m$ $$YSZ$ 3.34 \times 104 exp(-10300/T) $$/m$ $$/m$ $$/m$ $$YSZ$ $$Cathode support layer 760 $$ $$ $$ $$ $$ $$ $$ $$ $$ $$ $$ $$ $$$	Ni	$4.2 imes 10^{6} - 1065.3T$	S/m
$\begin{array}{llllllllllllllllllllllllllllllllllll$	Ionic conductivity		
YSZ $3.34 \times 10^4 exp(-10300/T)$ S/m Thickness mm Cathode support layer 760 µm (Ni + YSZ) mm (Ni + SSZ) Cathode active layer 10 µm (Ni + SSZ) mm (ScSZ) Anode active layer 15 µm (LSM + ScSZ) mm (LSM + ScSZ) Heat Conductivity V V/(m·K) Cathode 6.23 W/(m·K) Electrolyte 2.7 W/(m·K) Anode 9.6 W/(m·K) Heat Capacity V/(m·K) Heat Capacity Cathode 390 J/(kg·K) Electrolyte 300 J/(kg·K) Electrolyte 300 J/(kg·K) Electrolyte 2000 kg/m³ Anode 6870 kg/m³	ScSZ	$6.92 \times 10^4 exp(-9681/T)$	S/m
ThicknessCathode support layer760 μ m(Ni + YSZ) μ m μ m(Ni + ScSZ) μ mElectrolyte10 μ m(ScSZ) μ mAnode active layer15 μ m(LSM + ScSZ) μ m(LSM + ScSZ) μ mHeat Conductivity μ mCathode6.23 $W/(m.K)$ Electrolyte2.7 $W/(m.K)$ Electrolyte9.6 $W/(m.K)$ Heat Capacity U $J/(kg.K)$ Electrolyte300 $J/(kg.K)$ Electrolyte300 $J/(kg.K)$ Electrolyte2000 kg/m^3 Anode6870 kg/m^3	YSZ	$3.34 \times 10^4 exp(-10300/T)$	S/m
Cathode support layer (Ni + YSZ)760 μ m (Ni + YSZ)Cathode active layer (Ni + ScS2)10 μ m (ScSZ)Electrolyte (LSM + ScSZ)10 μ m (ScSZ)Anode active layer (LSM + ScSZ)15 μ m (Mr.K)Cathode Cathode6.23 $W/(m.K)$ (Mr.K)Electrolyte 2.72.7 $W/(m.K)$ (Mr.K)Electrolyte 2.79.6 $W/(m.K)$ (Mr.K)Heat Conductivity $U/(m.K)$ (Mr.K)Electrolyte 300 $J/(kg.K)$ 2.5Electrolyte 2.000 $M/(m.K)$ $MradeDensityUCathodeElectrolyte6870Electrolyte2.000kg/m^3Anode$	Thickness		
$\begin{array}{c c} ({\rm Ni} + {\rm YSZ}) & & & \\ \mbox{Cathode active layer} & 10 & & \mu m & \\ ({\rm Ni} + {\rm ScSZ}) & & & \\ \mbox{Electrolyte} & 10 & & \mu m & \\ ({\rm ScSZ}) & & & & \\ \mbox{Anode active layer} & 15 & & \mu m & \\ ({\rm LSM} + {\rm ScSZ}) & & & \\ \mbox{Heat Conductivity} & & \\ \mbox{Cathode} & 6.23 & & W/(m\cdot{\rm K}) & \\ \mbox{Electrolyte} & 2.7 & & W/(m\cdot{\rm K}) & \\ \mbox{Anode} & 9.6 & & W/(m\cdot{\rm K}) & \\ \mbox{Heat Capacity} & & \\ \mbox{Cathode} & 390 & & J/({\rm kg}\cdot{\rm K}) & \\ \mbox{Electrolyte} & 300 & & J/({\rm kg}\cdot{\rm K}) & \\ \mbox{Electrolyte} & 300 & & J/({\rm kg}\cdot{\rm K}) & \\ \mbox{Density} & & \\ \mbox{Cathode} & 6870 & & {\rm kg/m^3} & \\ \mbox{Electrolyte} & 2000 & {\rm kg/m^3} & \\ \mbox{Anode} & 6570 & & {\rm kg/m^3} \\ \end{array}$	Cathode support layer	760	μm
Cathode active layer 10 μ m (Ni + ScSZ) μ m Electrolyte 10 μ m (ScSZ) μ m (ScSZ) Anode active layer 15 μ m (LSM + ScSZ) μ m (ScSZ) Heat Conductivity 2.7 W/(m·K) Cathode 9.6 W/(m·K) Anode 9.6 W/(m·K) Heat Capacity U J/(kg·K) Electrolyte 300 J/(kg·K) Electrolyte 300 J/(kg·K) Density U Standa Cathode 6870 kg/m³ Electrolyte 2000 kg/m³ Anode 6570 kg/m³	(Ni + YSZ)		
$\begin{array}{c c c c } ({\rm Ni} + {\rm ScSZ}) & & & & & \\ \hline Electrolyte & 10 & & & & & \\ \mbox{(ScSZ)} & & & & & \\ \mbox{Anode active layer} & 15 & & & & & \\ \mbox{(LSM + ScSZ)} & & & & & \\ \hline Heat Conductivity & & & & \\ \mbox{(LSM + ScSZ)} & & & & \\ \hline Heat Conductivity & & & \\ \mbox{Cathode} & 6.23 & & & & & \\ \mbox{(m-K)} & & & \\ \mbox{Electrolyte} & 2.7 & & & & & \\ \mbox{Anode} & 9.6 & & & & & \\ \mbox{(m-K)} & & & \\ \mbox{Heat Capacity} & & & \\ \mbox{Cathode} & 390 & & & & & \\ \mbox{Lectrolyte} & 300 & & & & & \\ \mbox{Anode} & 420 & & & & & \\ \mbox{Anode} & 420 & & & & & \\ \mbox{Density} & & & & \\ \mbox{Cathode} & 6870 & & & & & & \\ \mbox{Electrolyte} & 2000 & & & & & \\ \mbox{Electrolyte} & 2000 & & & & & \\ \mbox{Belterolyte} & 2000 & & & & & \\ \mbox{Anode} & 6570 & & & & & \\ \end{tabular}$	Cathode active layer	10	μm
Electrolyte 10 μ m (ScSZ)	(Ni + ScSZ)		
(ScSZ) Anode active layer 15 μ m (LSM + ScSZ) (LSM + ScSZ) Heat Conductivity (LSM + ScSZ) Cathode 6.23 W/(m·K) Electrolyte 2.7 W/(m·K) Anode 9.6 W/(m·K) Heat Capacity (Lagracity) U Cathode 390 J/(kg·K) Electrolyte 300 J/(kg·K) Electrolyte 420 J/(kg·K) Density U U Cathode 6870 kg/m³ Electrolyte 2000 kg/m³	Electrolyte	10	μm
Anode active layer 15 μ m (LSM + ScSZ) (LSM + ScSZ) $Hat Conductivity Cathode 6.23 W/(m\cdot K) Electrolyte 2.7 W/(m\cdot K) Anode 9.6 W/(m\cdot K) Heat Capacity U V/(m\cdot K) Cathode 390 J/(kg\cdot K) Electrolyte 300 J/(kg\cdot K) Density U U Cathode 6870 kg/m^3 Electrolyte 2000 kg/m^3 $	(ScSZ)		
(LSM + ScS2) Heat Conductivity Cathode 6.23 W/(m·K) Electrolyte 2.7 W/(m·K) Anode 9.6 W/(m·K) Heat Capacity V V/(m·K) Cathode 390 J/(kg·K) Electrolyte 300 J/(kg·K) Density V V Cathode 6870 kg/m³ Electrolyte 2000 kg/m³ Anode 6570 kg/m³	Anode active layer	15	μm
Heat Conductivity Cathode 6.23 W/(m·K) Electrolyte 2.7 W/(m·K) Anode 9.6 W/(m·K) Heat Capacity V W/(m·K) Cathode 390 J/(kg·K) Electrolyte 300 J/(kg·K) Danode 420 J/(kg·K) Density V Kg/m³ Electrolyte 2000 kg/m³ Anode 6570 kg/m³	(LSM + ScSZ)		
Cathode 6.23 $W/(m\cdot K)$ Electrolyte 2.7 $W/(m\cdot K)$ Anode 9.6 $W/(m\cdot K)$ Heat Capacity $V/(m\cdot K)$ $V/(m\cdot K)$ Cathode 390 $J/(kg\cdot K)$ Electrolyte 300 $J/(kg\cdot K)$ Anode 420 $J/(kg\cdot K)$ Density V $V/(m \cdot K)$ Electrolyte 2000 kg/m^3 Anode 6570 kg/m^3	Heat Conductivity		
Electrolyte 2.7 $W/(m\cdot K)$ Anode 9.6 $W/(m\cdot K)$ Heat Capacity $V/(m\cdot K)$ $V/(m\cdot K)$ Cathode 390 $J/(kg\cdot K)$ Electrolyte 300 $J/(kg\cdot K)$ Anode 420 $J/(kg\cdot K)$ Density V V Cathode 6870 kg/m^3 Electrolyte 2000 kg/m^3	Cathode	6.23	W/(m·K)
Anode 9.6 W/(m·K) Heat Capacity Cathode 390 J/(kg·K) Electrolyte 300 J/(kg·K) Anode 420 J/(kg·K) Density Cathode 6870 kg/m³ Electrolyte 2000 kg/m³ Anode 6570 kg/m³	Electrolyte	2.7	W/(m·K)
Heat Capacity Cathode 390 J/(kg·K) Electrolyte 300 J/(kg·K) Anode 420 J/(kg·K) Density Zathode 6870 kg/m³ Electrolyte 2000 kg/m³ Anode 6570 kg/m³	Anode	9.6	W/(m·K)
Cathode 390 J/(kg·K) Electrolyte 300 J/(kg·K) Anode 420 J/(kg·K) Density	Heat Capacity		
Electrolyte 300 J/(kg·K) Anode 420 J/(kg·K) Density	Cathode	390	J∕(kg⋅K)
Anode 420 J/(kg·K) Density	Electrolyte	300	J∕(kg⋅K)
DensityCathode6870kg/m³Electrolyte2000kg/m³Anode6570kg/m³	Anode	420	J∕(kg⋅K)
Cathode 6870 kg/m³ Electrolyte 2000 kg/m³ Anode 6570 kg/m³	Density		
Electrolyte 2000 kg/m³ Anode 6570 kg/m³	Cathode	6870	kg/m ³
Anode 6570 kg/m ³	Electrolyte	2000	kg/m ³
	Anode	6570	kg/m ³

Table 2

Material parameters of Co₃O₄ and CoO[34].

Parameter	Value or expression	Unite
Heat Capaci	ţy	
Co ₃ O ₄	0.55T + 328.6	J/(kg·K)
CoO	0.12T + 628	J/(kg·K)
Heat Conduc	tivity	
Co ₃ O ₄	$-1.25 \bullet 10^{-8}T^3 + 4.27 \bullet 10^{-5}T^2 - 5.78 \bullet 10^{-2}T + 33.381$	W∕ (m⋅K)
CoO	_	. ,
Density		
Co ₃ O ₄	6110	kg/m ³
CoO	6450	kg/m ³

absorbed or supplied by the TES sub-model. Fig. 1 depicts the working schematic. The multi-physics model is calculated numerically by the finite element method.



2.1. SOEC sub-model

The electrochemical reaction rates of steam and carbon dioxide are calculated in this part. Driven by the operating voltages, the electrolysis reactions occur in the porous electrodes can be written as Eqs. (1)-(5), respectively.

In the porous cathode:

$$2H_2O + 4e^- \rightarrow 2H_2 + 2O^{2-}$$
 (1)

$$2\mathrm{CO}_2 + 4\mathrm{e}^- \rightarrow 2\mathrm{CO} + 2\mathrm{O}^2^- \tag{2}$$

In the porous anode:

$$20^2 \rightarrow O_2 + 4e^- \tag{3}$$

The overall electrochemical reactions:

$$2H_2O \rightarrow 2H_2 + O_2 \tag{4}$$

$$2CO_2 \rightarrow 2CO + O_2 \tag{5}$$

The equilibrium voltage and local electrochemical reaction rate for

Table	3	

Parameters	Value	Unit
Tortuosity		
Cathode	3	
Anode	3	
Electrochemical kinetics		A/m ²
H ₂	$3.3 imes10^8$	
O ₂	$4.2 imes10^7$	
Charge transfer coefficient		
H ₂	0.65	
CO	0.65	
02	0.5	

Table 4	4
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Operating parameters for the study of effects of anode channel width.

Parameter	Value	Unite
Operating potential	0.95–1.5	v
Cathode total gas utilization	60 %	
Inlet gas temperature	1123	K
Inlet H ₂ O mole fraction	0.6	
Inlet CO ₂ mole fraction	0.4	
Air flow rate	1000	SCCM
Anode channel width	2, 3, 4, 5	mm
Initial conversion of Co ₃ O ₄	0.5	





Fig. 2. (a) Model validation for SOEC sub-model [30], (b) profile of fluctuating voltage input to SOEC.



Fig. 3. Dynamic performance of the (a) Average temperature, (b) conversion of Co₃O₄, (c) current density, and (d) H₂/CO ratio at different anode channel width.

co-electrolysis can be determined by Nernst equation and Butler-Volmer equation, respectively.

Nernst equation:

$$E_{eq,H_2O} = 1.253 - 0.00024516T + \frac{RT}{2F} \ln \left[\frac{P_{H_2}^C \left(P_{O_2}^A \right)^{1/2}}{P_{H_2O}^C} \right]$$
(6)

$$E_{eq,CO_2} = 1.46713 - 0.0004527T + \frac{RT}{2F} ln \left[\frac{P_{CO}^{C} \left(P_{O_2}^{A} \right)^{1/2}}{P_{CO_2}^{C}} \right]$$
(7)

Where R is the ideal gas constant, T is the local temperature (K), $P_{H_2}^C$, $P_{H_2O}^C$, P_{CO}^C , and $P_{CO_2}^C$ are the partial pressure of H_2 , H_2O , CO and CO₂ at the reaction sites in the cathode; $P_{O_2}^A$ is the partial pressure of O₂ at the reaction sites in the anode. F is the Faraday constant.

It can be noted that the local gas partial pressures are included in Eqs. (6)–(7). Therefore, the concentration overpotential is included in the Nernst potential calculation.

Butler-Volmer equation is applied to calculate the activation overpotential (η_{act}) at a given current density (i_{H_2O} for steam electrolysis):

$$i_{H_2O} = \beta_{H_2} \frac{P_{H_2O}}{P_{ref}} \frac{P_{H_2}}{P_{ref}} \exp(-\frac{Ea}{RT}) \left\{ \exp\left(\frac{\alpha n F \eta_{act}}{RT}\right) - \exp\left(-\frac{(1-\alpha) n F \eta_{act}}{RT}\right) \right\}$$
(8)

Where the β_{H_2} value is $3.3 \times 10^8 (\text{A/m}^2)$, which is about 2.2 times larger than that of CO [30]. n is the number of electrons transferred per reaction. E is the activation energy (J/mol). α is the symmetrical factor and is usually taken as 0.5 for SOEC/SOFC.

At the porous cathode, in addition to electrochemical reactions, water gas shift reaction (WGSR) is considered as an important reaction to balance the concentration of H_2O , H_2 , CO_2 and CO:

$$CO + H_2O \leftrightarrow CO_2 + H_2$$
 (9)

The reaction rate of WGSR is greatly increased with Ni catalyst (at the cathode area) and can be neglected in the absence of Ni, which can be calculated by the widely used expressions as [31]:

$$R_{WGSR} = k_{sf} (P_{H_2 O} P_{CO} - \frac{P_{H_2} P_{CO_2}}{K_{ps}})$$
(10)

$$k_{sf} = 0.0171 \exp(\frac{-103191}{RT}) \tag{11}$$

$$K_{ps} = \exp(0.3169 + 4.1788Z + 0.6351Z^2 - 0.2935Z^3)$$
(12)

$$Z = \frac{1000}{T} - 1$$
(13)

The local heat source in SOEC (Q_{SOEC}) is defined as:

$$Q_{SOEC} = Q_{act} + Q_{ohmic} + Q_{ec} + Q_{WGSR}$$

$$\tag{14}$$

Where Q_{act} is the activation loss related to energy barrier for electrochemical reaction (Eq. (15)). Q_{ohmic} is the Ohmic loss caused by the resistance to ionic/electronic conduction (Eq. (16)). Q_{ec} is the entropy change ($\Delta S:J/(\text{mol-K})$) of the H₂O and CO₂ reduction reactions and can be calculated by Eq. (17). Q_{WGSR} is the enthalpy change of WGSR and related to the reaction rate (Eq. (18)).

$$Q_{act} = |\eta_{act}| \cdot i \tag{15}$$



Fig. 4. (a) & (b) Breakdowns of heat from different process in the cell, (c) & (d) diagrams of temperature gradient in length direction at the interface of cathode and electrolyte, (e) & (f) temperature distribution through the cell surface.

Table 5

Operating parameters for the study of effects of initial conversion of Co₃O₄.

Parameter	Value	Unite
Operating potential	0.95-1.5	v
Cathode total gas utilization	60 %	
Inlet gas temperature	1123	K
Inlet H ₂ O mole fraction	0.6	
Inlet CO ₂ mole fraction	0.4	
Air flow rate	1000	SCCM
Anode channel width	4	mm
Initial conversion of Co ₃ O ₄	0.2, 0.5, 0.8	

$$Q_{ohmic} = \frac{i_s^2}{\sigma_s} + \frac{i_l^2}{\sigma_l}$$
(16)

$$Q_{ec} = \Delta S \frac{T \cdot i}{n \cdot F} \tag{17}$$

$$Q_{WGSR} = \Delta H \cdot R_{WGSR} \tag{18}$$

The properties of electrode and electrolyte such as geometry, ionic, electronic conductivity and thermodynamic parameters can be found in Table 1. Porosity and active specific surface area are calculated by a



Fig. 5. Dynamic performance of the average temperature at different initial conversion rates of Co_3O_4 .

combination of experiment and permeation theory, with values of 0.36 and $2.14 \times 10^5 (1/m)$, respectively, for porous electrode. A more complete discussion of the electrochemical equations can be found in our previous works [32].

2.2. TES sub-model

In this paper, Co_3O_4/CoO redox pair is chosen as the TES material, whose redox reaction temperature region is in the range of 800 ~ 900 °C with good reaction kinetics and cyclic stability. The reduction process of Co_3O_4 is an endothermic reaction:

$$Co_3O_4 \rightarrow 3CoO + 0.5O_2\Delta H_{298,15K} = 196.2kJ/mol$$
 (19)

The oxidation process of CoO is an exothermic reaction:

$$3C_{0}O + 0.5O_{2} \rightarrow C_{03}O_{4}\Delta H_{298.15K} = -196.2kJ/mol$$
 (20)

Pagkoura et al. [33] tested the reaction rate and developed the kinetic model, which was well validated in the numerical simulations of Singh et al. [34] and Yang et al. [35] as follows:

$$R_{Co_3O_4} = k_{re} \cdot (1 - X) - k_{ox} \cdot X$$
(21)

Table 6

Operating parameters for the study of effects of inlet gas temperature.

Parameter	Value	Unite
Operating potential	0.95–1.5	V
Cathode total gas utilization	60 %	
Inlet gas temperature	1103, 1123, 1143	K
Inlet H ₂ O mole fraction	0.6	
Inlet CO ₂ mole fraction	0.4	
Air flow rate	1000	SCCM
Anode channel width	4	mm
Initial conversion of Co ₃ O ₄	0.5	



Fig. 6. (a) Thermochemical reaction rate distribution, (b) breakdown of heat from different process, (c) temperature gradient in length direction at the interface of cathode and electrolyte and (d) temperature distribution at 8:40.



Fig. 7. Dynamic performance of the average temperature at different inlet temperatures.

Where the k_{re} and k_{ox} is the rate constant of reduction and oxidation reaction, respectively:

$$k_{re} = 2.12 \cdot 10^{27} \cdot e^{-\frac{E_{re}}{T}}$$
(22)

$$k_{\rm ex} = 1.4 \cdot 10^{-15} \cdot e^{-\frac{E_{\rm ex}}{T}} \tag{23}$$

Where the $E_{re} = 87000$ K and $E_{ox} = 30000$ K. X represents the conversion of Co₃O₄,

$$X = 1 - \frac{n_{Co_3O_4}}{n_{Co_3O_{40}}}$$
(24)

Where $n_{Co_3O_4}$ represents the current concentration of Co₃O₄, and $n_{Co_3O_{40}}$ represents the initial concentration.

The heat source of TES (Q_{TES}) can be calculated as:

$$Q_{TES} = -R_{Co_3O_4} \cdot \Delta H \tag{25}$$

TES material is assumed as a porous media with a fixed porosity of 0.7 to ensure a good gas transport. And the material properties of Co_3O_4 and CoO are listed in Table 2.

2.3. CFD sub-model

In the CFD sub-model, the changes in physical variables due to SOEC processes and TES processes are coupled to the mass conservation (Eq. (26)) and energy conservation (Eq. (28)) equations using source terms. Mass conservation equation:

$$\rho \frac{\partial \rho}{\partial t} + \nabla(\rho u) = M_{\text{source}}$$
(26)

Gas diffusion:

$$N_i = -\frac{1}{RT} \left(\frac{B_0 y_i P}{\mu} \frac{\partial P}{\partial z} - D_i^{\text{eff}} \frac{\partial (y_i P)}{\partial z} \right) (i = 1, ..., n)$$
(27)

Energy conservation:

$$(\rho C_p)_{eff} \frac{\partial T}{\partial t} + \rho C_{p,g} U \nabla T + \nabla \cdot \left(-\lambda_{eff} \nabla T \right) = Q_{\text{source}}$$
(28)

Where the $\lambda_{e\!f\!f}$ is the effective thermal conductivity and can be calculated as:

$$\lambda_{eff} = (1 - \varepsilon)\lambda_s + \varepsilon\lambda_l \tag{29}$$

The incompressible momentum conservation equation in the gas channels:

$$\rho \frac{\partial \mathbf{u}}{\partial \mathbf{t}} + \rho \mathbf{u} \nabla \mathbf{u} = -\nabla \mathbf{p} + \nabla \mu \left[\left(\nabla \mathbf{u} + \left(\nabla \mathbf{u} \right)^T \right) - \frac{2}{3} \mu \nabla \mathbf{u} \right]$$
(30)

Particularly, in the porous electrodes and TES material, the equation is further corrected by introducing the Darcy's Law term:

$$\frac{\partial \mathbf{u}}{\partial t} + \rho \mathbf{u} \nabla \mathbf{u} = -\nabla \mathbf{p} + \nabla \mu \left[\left(\nabla \mathbf{u} + (\nabla \mathbf{u})^T \right) - \frac{2}{3} \mu \nabla \mathbf{u} \right] - \frac{\varepsilon \mu \mathbf{u}}{\kappa}$$
(31)

Ideal gas assumption and local thermal equilibrium assumption are adopted in this model. A detailed description of the mentioned control equations can be found in [32].

2.4. Boundary conditions

2.4.1. Mass conservation

The gas composition is defined at the inlets of the two channels, and the convective flux boundary condition is defined at the outlets of the two channels. The continuity boundary condition is specified at the electrode/channel interfaces. At other boundaries, zero flux is applied:

$$-n \cdot N = 0 \tag{32}$$

2.4.2. Momentum conservation

Gas flow rate is given at the inlets of the two channels, and standard atmospheric pressure is set at the outlet (Eq. (33)). No slip condition is applied to the electrolyte/electrode interface and the ends of electrodes (Eq. (34)).

$$P_{out} = 0 \tag{33}$$

$$u_w = 0 \tag{34}$$

2.4.3. Energy conservation

The inlet gas temperature is defined at the inlets of the two channels and the convective heat flux boundary is defined at the outlets. The cell's outside wall is an adiabatic boundary:

$$-n \cdot q = 0 \tag{35}$$

2.4.4. Electrochemical reaction

The working potential ($V_{cell,ca}$) and the electrical ground ($V_{cell,an}$) are set on the outside of the cathode and anode, respectively, and the remaining boundaries are electrically insulated (Eq. (38)).

$$V_{cell,ca} = \phi_{working} \tag{36}$$

$$V_{cell,m} = 0 \tag{37}$$

$$-n \cdot i = 0 \tag{38}$$

2.5. Model validation and input profile

As can be seen in Fig. 2a, the simulation results of the SOEC submodel agree well with the experimental results [30] under 1023 K/ 973 K and a certain cathode inlet gas composition (28.6 % $CO_2 + 28.6$ % $H_2O + 14.3$ % $H_2 + 28.6$ % Ar), which validates the feasibility of the model. The values of turning parameters are set in accordance with the principle to minimize the deviation between simulation results and experimental results at two temperature cases. As the preferred operating temperature is 1123 K in this work, the parameters are tuned to fit better with the higher temperature case as listed in Table 3. As the kinetic parameters and material properties are all functions of temperature, we believe the validated model can be further extended to wider temperature ranges. As for the extension of operating voltage, once kinetic parameters of the electrochemical reaction are validated, operating voltage can also be extrapolated according to the widely used Butler-Volmer equation.



Fig. 8. (a) & (b) Thermochemical reaction rate distribution, (b) & (c) breakdown of heat from different process, (e) & (f) temperature gradient in length direction at the interface of cathode and electrolyte.

 Table 7

 Operating parameters for the study of effects of cathode inlet gas composition.

Parameter	Value	Unite
Operating potential	0.95–1.5	V
Cathode total gas utilization	60 %	
Inlet gas temperature	1123	K
Inlet H ₂ O mole fraction	0.4, 0.6, 0.8	
Inlet CO ₂ mole fraction	0.6, 0.4, 0.2	
Air flow rate	1000	SCCM
Anode channel width	4	mm
Initial conversion of Co ₃ O ₄	0.5	

In order to evaluate the heat management effects of TES when renewable energy is used as an input, a real-time PV power output profile during a sunny day around the vernal equinox day in Zhejiang Province China is chosen as the fluctuating voltage input as shown in Fig. 2b. To balance its production rate and economic feasibility, we select 1.5 V as the maximum operation voltage and 0.95 V as the minimum operating voltage (a value close to the Nernst potentials in the case studies) [7].



Fig. 9. (a) & (b) Dynamic performance of the cell, (c) & (d) temperature gradient in length direction at the interface of cathode and electrolyte.

3. Results and discussion

3.1. Effects of anode channel width

The heat absorbed and released by the TES is related to the filling quantity, which is determined by the anode channel width with a fixed fill porosity. Therefore, the anode channel width is vital to the thermal characteristics of the cell. The width of the air channel is usually 1-2 mm, which is extended to 2, 3, 4, 5 mm to study the effect of the filling quantity. A high gas utilization rate is desired in the practical operation of SOEC, but a thoroughly utilization will cause "reactant starvation". To ensure the long-term safety and stability in operation, a gas utilization rate of 60 % is selected. A high air flow rate can timely purge the oxygen generated by the electrochemical reaction, reducing the partial pressure of oxygen, and can also play a role in regulating the temperature. However, a high air flow rate requires additional energy costs. For our model, an air flow rate of 1000 SCCM is appropriate. The inlet gas temperature and conversion of Co₃O₄ need to be properly set as they are key factors on the kinetics of the thermochemical reactions, which will be discussed in more detail later. The operating conditions in this section are given in Table 4.

The temperature fluctuates along with the voltage variation, where TES can significantly reduce the fluctuation amplitude, as shown in Fig. 3a. Without TES, the daytime temperature difference is approximately 119.2 K, which is more than twice as much as the 2-channel-width case with TES. In the meanwhile, the decrease of anode channel width contributes to a larger effect of TES on the temperature field. The conversion of Co_3O_4 in the 5-channel-width case is changed by 35 % and 38 % in endothermic and exothermic stages, respectively, which are

increased to 62 % in the 2-channel-width case. This means that a large part of TES is not utilized with the increase of anode channel width. Moreover, increasing the anode channel width will increase the cell volume and reduce the energy density, indicating that it is unreasonable to increase the anode channel width infinitely. The small difference in mole fraction of O₂ among different TES filling amounts indicates that the changes of oxygen concentration caused by redox reactions of TES materials can be ignored, as shown in Fig. 3b. TES can significantly reduce temperature and improve cell safety at high voltages, but also impairs the current density as shown in Fig. 3c. At 11:00, the current density of 2-channel-width case reaches about 3.4 A/cm^2 , which is 35 %lower than that without TES. Fig. 3d indicates the combination of TES has little effect on the H₂/CO ratio of the output gas.

Breakdowns of heat sources at two typical stages (8:40-endothermic stage & 11:00-exothermic stage) are further presented in Fig. 4a & b. The heat released/consumed from the thermochemical reactions both increases with the anode channel width at 8:40 and 11:00, which well explains the temperature variation in Fig. 3a. As further illustrated in Fig. 4c, the largest temperature gradient (-3.9 K/cm) appears in the inlet area of the cell at low voltages without TES. As the anode channel width increases, the largest temperature gradient becomes positive and migrates towards the back of the cell. Without TES, the maximum temperature gradient at the inlet reaches 74.7 K/cm at 11:00, which is 177 % higher than that at 4-channel-width case, as shown in Fig. 4d. In the middle of the cell, the temperature gradient is only 1.5 K/cm at 4-channel-width case, which is 92 % lower than that without TES. It is indicated that thermochemical heat storage material can effectively reduce temperature gradients at high voltages. The introduction of TES not only helps to inhibit temperature fluctuation throughout the day, but also reduces the temperature difference across a single cell surface. The temperature distribution of two typical nodes (8:40 & 11:00) is shown in Fig. 4e & 4f. With TES, the temperature distribution on the whole cell surface becomes more uniform, further confirming the results in Fig. 4c and Fig. 4d. With the increase in width, the maximum temperature occurs at the outlet instead of the inlet at 8:40, and the maximum temperature difference is about 10 K at 4-channel-width case, which is 49 % lower than that without TES. At 11:00, the maximum temperature difference is 168 K without TES, while it is only 30 K (82 % lower) at 4-channel-width case.

3.2. Effects of initial conversion of Co_3O_4

The kinetic parameters of TES are related to the conversion of Co_3O_4 . Therefore, different initial conversions are examined to determine their influence on the temperature during the start-up early in the morning. In this section, we investigate the effect of the initial conversion of Co_3O_4 , and the operating parameters are presented in Table 5.

The initial temperature increases with the increase of the initial conversion of Co_3O_4 , as seen in Fig. 5. At 6:40, the average temperature of 0.8-conversion case reaches about 1154 K, which is approximately 28.4 K higher than that of 0.2-conversion case. However, a higher or lower initial temperature can also result in a larger temperature fluctuation. The temperature standard deviations for 0.8-conversion, 0.5-conversion and 0.2-conversion are 10.28, 8.51, and 8.56, respectively. Meanwhile, it is found that different initial conversions have little effect on the endothermic stage at noon and the exothermic stage in the afternoon.

Fig. 6a illustrates the thermochemical reaction rate distribution at 8:40. The absolute value of the reaction rate increases along the length of the cell and increases with the initial conversion of Co_3O_4 . The higher the reaction rate, the more heat produced from the thermochemical reaction (Fig. 6b), which in turn increases the SOEC temperature. As the initial conversion increases, the cell changes from endothermal operating condition to exothermal operating condition, and the maximum temperature gradient (4.7 K/cm) occurs at 0.8-conversion case, as shown in Fig. 6c. Similarly, the maximum temperature difference across the surface increases with the initial conversion (Fig. 6d).

3.3. Effects of inlet gas temperature

Operating temperature is a key factor on the kinetics of the thermochemical and electrochemical reactions, which will affect the temperature distribution inside the cell. The effects of inlet gas temperature are discussed in this section, as well as the associated operating parameters are presented in Table 6.

As can be seen in Fig. 7, the 1123 K case presents the lowest temperature fluctuation with a standard deviation of 8.51, while the values at 1103 K and 1143 K are 12.56 and 9.65, respectively. It can also be found from Fig. 7 that the average temperature is close to the gas inlet temperature during startup at 1143 K case. This is due to the fact that at 0.5 conversion rate of Co₃O₄, 1143 K nearly exceeds the exothermic temperature of Co₃O₄/CoO redox pair, resulting in a slow oxidation reaction rate (Fig. 8a). Therefore, the heat production from thermochemical reaction is the lowest, as shown in Fig. 8c. A higher temperature is favorable for the reduction reaction. At 11:00, the fastest reduction reaction rate occurs at 1143 K case, as shown in Fig. 8b. As a result, the heat consumption from thermochemical reaction is the highest (Fig. 8d). However, a higher temperature will also cause greater over-potential losses. In contrast, the heat absorption from TES is insufficient to maintain the cell temperature (Fig. 7). Overall, an inlet temperature of 1123 K is optimal to match the electrochemical and thermochemical kinetics.

The temperature gradients along the flow channel at the interface between cathode and electrolyte (8:40 & 11:00) are further presented in Fig. 8e & 8f. For comparison, the temperature gradients of each different inlet gas temperatures without TES are also shown in the Fig. 8e & 8f. At 8:40, the largest temperature gradient is close to the case without TES, while it decreases by 64 % to 30.5 K/cm at 11:00.

3.4. Effects of cathode inlet gas composition

The electrochemical activities of H_2O and CO_2 are very different and there also exists water gas shift reaction that affects the conversion among these reactants which have a significant effect on SOEC dynamic behavior. Therefore, the effects of cathode inlet gas composition are studied in this section, and the detailed operating parameters are shown in Table 7.

As shown in Fig. 9a, a low H₂O mole fraction can help inhibit temperature fluctuation, where the maximum daytime temperature difference is 22.6 K at 0.4-H₂O case. However, this will reduce the output H₂/CO ratio, as shown in Fig. 9b. It is recommended to operate with an inlet H₂O mole fraction of 0.6 to maintain H₂/CO ratio > 2, which is favorable for downstream chemical synthesis processes such as Fischer-Tropsch synthesis. When the TES is coupled to the SOEC, the maximum temperature gradient (3.3 K/cm) occurs at 0.4-H₂O case in the morning endothermic stage in Fig. 9c, whose absolute value is also close to that without TES. As the operating voltage increases, the maximum temperature gradient increases to 34 K/cm near the inlet at 0.8-H₂O case (Fig. 9d), which is 61 % lower than that without TES. It is worth noting that the temperature gradient at the rear of the cell does not differ significantly for different inlet H₂O mole fraction cases with TES at high voltages.

4. Conclusion

In summary, we have proposed a novel strategy for the adaptive heat management of SOEC by coupling with an in-situ thermochemical energy storage (TES) layer. We find the combination of TES section can effectively suppress temperature fluctuation, where the temperature difference during the day is significantly reduced from 119.2 K to 31.7 K at a TES width of 4 mm. The maximum temperature gradient and the surface maximum temperature difference at 1.5 V are also reduced to 26.2 K/cm and 30 K, respectively, which are 65 % and 82 % lower than those without TES. The lowest fluctuation rate occurs in the 0.5-conversion case with the standard deviation of average temperature being 8.51. An inlet temperature of 1123 K appears as the optimal to match electrochemical and thermochemical kinetics, where the lowest average temperature fluctuation can be achieved. The addition of TES can also stabilize the temperature gradient at different inlet H₂O mole fractions, and an inlet H₂O mole fraction of 0.6 is recommended to maintain an H₂/CO ratio above 2. This work demonstrates the feasibility of integrating thermochemical reaction section with SOEC for flexible heat management of SOEC with fluctuating power supplies.

CRediT authorship contribution statement

Anwei Sun: Data curation, Methodology, Software, Validation, Formal analysis, Writing – original draft, Writing – review & editing. Wei Shuai: Writing – review & editing. Nan Zheng: Writing – review & editing. Yu Han: Writing – review & editing. Gang Xiao: Funding acquisition, Investigation, Resources, Supervision, Writing – review & editing. Meng Ni: Funding acquisition, Writing – review & editing. Haoran Xu: Conceptualization, Project administration, Methodology, Supervision, Formal analysis, Resources, Funding acquisition, Writing review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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