The development of solid oxide co-electrolysis of H_2O and CO_2 on large-size cells and stacks

Jingjing Liang[1], Jianzhong Zhu[1], Minfang Han[1] ✉, Xiufu Hua[2], Duruo Li[2] and Meng Ni[3]

ABSTRACT

In the context of carbon neutrality, conversion of CO_2 into CO is an effective way for negative carbon emission. Electrochemical reduction is a novel developed pathway, among which, solid oxide co-electrolysis technology is promising for its high efficiency and low electricity demand. Researches concerning the large-size cell and stack of application level are important. This review, targeting at the not yet fully understood reaction mechanism and the most concerning issue of durability, details the reported factors playing important roles in the reaction mechanism and durability of co-electrolysis. It is found that the operating conditions such as inlet mixtures and applied current significantly affect the reaction mechanism of co-electrolysis and the experiments on button cells can not reflect the real reaction mechanism on industrial-size cells. Besides, the durability test of large-size single cells and stacks at high current with high conversion rate and the potential of solid oxide co-electrolysis combing with intermittent renewable energy are also reviewed and demonstrated. Finally, an outlook for future exploration is also offered.

KEYWORDS

Solid oxide co-electrolysis, large-size cell, stack, reaction mechanism, durability.

Fossil fuel as a major energy source helps fulfill the energy demand of the development of modern society, but at the same time the related large amount of CO_2 emission and its concomitant environmental issue have elicited the attention around the world. In 2020, China announced the vision of carbon peak and carbon neutrality targets based on the inherent requirement to promote sustainable development. In this context, the demand for technological innovations in carbon capture and utilization (CCU) is gradually increasing and researches concerning not only a transition to alternative environment-friendly energy source but also mitigation and repurposing of excessive CO_2 are in full swing[1, 2]. CO_2 can be converted to a wide range of add-value chemicals such as synthetic fuels etc. and the routes have been grouped in detail in Ref. [1] as showed in Figure 1.

Among these routes, electrochemical reduction of CO_2 has recently picked up a lot of ground for the advantages of its high reaction rate, controllable conversion rate through electrode potential, tunable product selectivity, clean power source such as solar and wind renewable energy, and high potential for large scale application[3–6]. Compared with other low temperature and medium temperature electrolysis technologies, solid oxide electrolysis cell (SOEC) technology emerges as the leader in terms of Faradaic efficiency, energetic efficiency and electric power consumption for CO_2 conversion[7, 8]. Compared with alkaline electrolysis (AE) and proton exchange membrane electrolysis (PEME) with relatively higher technology readiness, SOEC can directly convert CO_2 into CO while the others are not designed to. Besides, the high temperature (600–900 °C) operation reduces the demand for electricity and enables the utilization of excess heat from nuclear power plant, metallurgical plant, etc., which would significantly improve the system efficiency.

However, the attempts to convert CO_2 through SOEC were mainly conducted in the form of co-electrolysis, in which case both H_2O and CO_2 were introduced into the cell simultaneously and syngas can be obtained[9, 10]. Generally, the performance of CO_2 electrolysis was significantly inferior to that of H_2O electrolysis because of the lower diffusion rate of CO_2 and higher activation energy of CO_2 reduction. In comparison to pure CO_2 electrolysis, co-electrolysis showed a lower activation energy, lower polarization resistance, and faster electrochemical kinetic[10–12]. Besides, carbon deposition which would cause irreversible damage to the fuel electrode can be suppressed when H_2O was injected along with CO_2. What's more, syngas with tunable ratio of H_2/CO can be obtained in one reactor and can be utilized for downstream sectors[13, 14].

However, the reaction mechanism is not yet fully understood for co-electrolysis. To estimate the energy requirement, composition and production rate of syngas, the reaction mechanism, that is, the reaction processes should be clarified[10]. Besides, satisfactory performance of co-electrolysis was constantly demonstrated in recent years on either button cells or stacks, and the performance was no longer the major obstacle in the commercialization of SOEC[15–17]. Durability and longevity have been the main limiting factors. Studies on co-electrolysis were mostly based on the state-of-the-art Ni-yttria-stabilized zirconia (Ni-YSZ) cermet supported cells. Some other alternative materials were also explored and applied, but Ni-YSZ fuel electrode was overwhelming in industrial application[18]. To understand the development status of SOEC co-electrolysis technology for future application, this review mainly focused on the reaction mechanism and durability of SOEC co-electrolysis with conventional Ni-YSZ fuel electrode on large-size single cells and stacks.

[1]Fuel Cell and Energy Storage Center, Department of Energy and Power Engineering, Tsinghua University, Beijing 100084, China; [2]Yangtze Delta Region Institute of Tsinghua University, Zhejiang, Jiaxing 314006, China; [3]Department of Building and Real Estate, The Hong Kong Polytechnic University, Hong Kong 999077, China
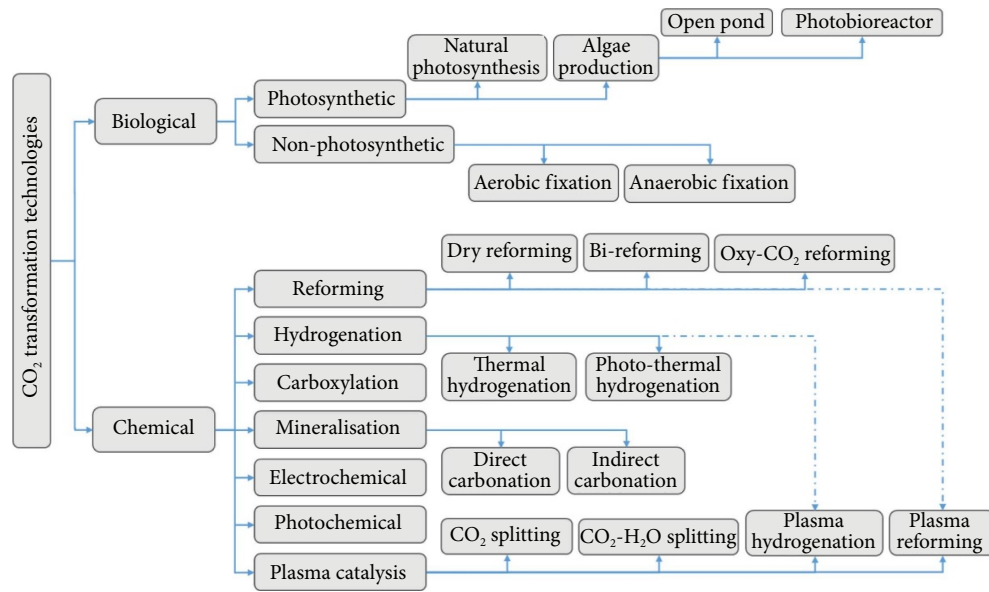Address correspondence to Minfang Han, hanminfang@tsinghua.edu.cn

Fig. 1   Classification of CO_2 utilization technologies. Reprinted with permission from Ref. [1], © 2021 Elsevier.

1   Reaction mechanism of co-electrolysis on large-size single cells

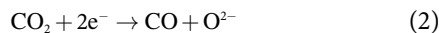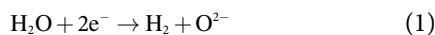## 1.1   Description of solid oxide co-electrolysis processes

The working principle of solid oxide co-electrolysis was shown in Figure 2.

Mixture of $CO_2+H_2O+H_2$ or CO_2+H_2 was always injected into the cathode of SOEC, where H_2 was indispensable for Ni-based cathode protection. Syngas with tunable ratio of H_2/CO can be acquired in the outlet. Meanwhile, O_2 was produced at the side of anode.

The possible reactions occurred during co-electrolysis were showed as follows:

Cathode:

$$H_2O + 2e^- \rightarrow H_2 + O^{2-} \tag{1}$$

$$CO_2 + 2e^- \rightarrow CO + O^{2-} \tag{2}$$

$$H_2 + CO_2 \leftrightarrow H_2O + CO \, (\text{RWGS reaction}) \tag{3}$$

Anode:

$$O^{2-} - 2e^- \rightarrow \frac{1}{2}O_2 \tag{4}$$

For sole H_2O electrolysis and sole CO_2 electrolysis, only Equations (1) and (2) occurred respectively. In the case of co-electrolysis, both Equations (1) and (2) were possible to proceed simultaneously. There may be competition between these two reactions. Except for electrochemical reaction, the reversible thermochemical reaction of water gas shift (RWGS) reaction was also parallel, thus making the real reaction processes during co-electrolysis complicated. The fact of easier splitting of steam due to the lower activation energy and faster diffusion triggered the discussion about whether CO_2 electrochemical splitting still occurred in the presence of steam already. And if CO_2 electrochemical reduction did occur, what was the contribution of CO_2? Bisides, how did the operating conditions (injecting mixtures, applied current, etc.) affect the contribution of these two reactions? These questions were con-
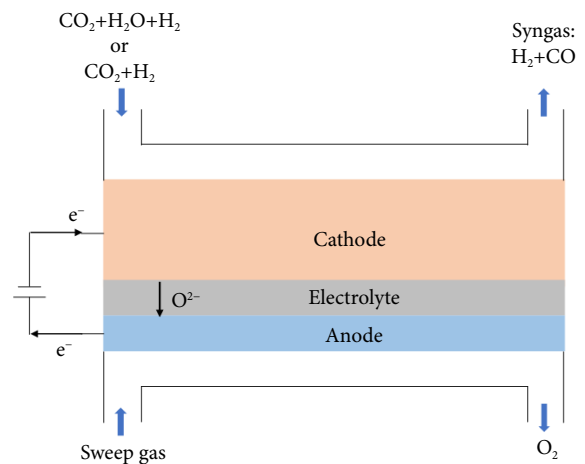

Fig. 2   working principle of solid oxide co-electrolysis.

stantly answered and discussed by various research groups all over the world.

1.2   Reaction mechanism differences based on button cells and large-size cells

However, so far, there has been no consensus about the reaction mechanism of co-electrolysis. One of the main reasons was that the relevant exploration was mainly conducted on button cells[19–22] and only limited results were reported on large-size single cells ($\geqslant$ 16 cm², usually 100 cm²)[10, 23–28]. The button cells could not reflect the gas composition and current density unevenness, etc., of large-size cells[29, 30] because of a low reactant conversion rate and an electrode surface-perpendicular gas supply path. This resulted in that the reaction mechanism explored on button cells could not represent the real reaction mechanism of large-size cells. For example, our group[22] reported the dominant electrolysis of H_2O during co-electrolysis on a button cell, while, it was meanwhile reported that the CO_2 electrolysis dominated the performance of co-electrolysis when the applied current density was higher than 0.13 A·cm⁻² on a 100 cm² industrial-size cell[23] when the same mixture of 90% CO_2+ 10% H_2 was supplied as showed in Figure 3.

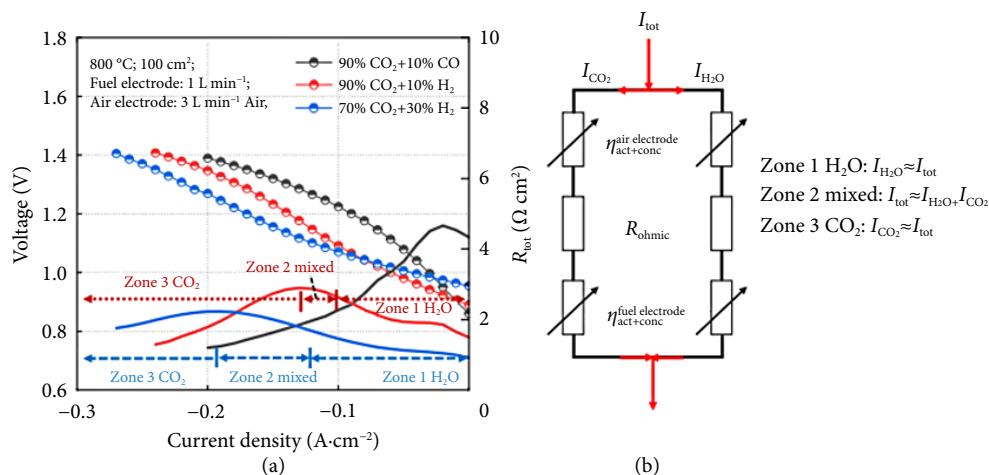That was, when the button cell and the industrial-size cell sharing

Fig. 3 (a) The effect of applied current and inlet mixture on the reaction mechanism of co-electrolysis; (b) the possible equivalent circuit of co-electrolysis[23].

a similar structure were operated at the same operating conditions, different reaction mechanisms were reflected. The performance near the inlet part of a large-size cell can be simulated by a button cell with the same structure at the same operating conditions. The different reaction mechanism of the button cell and the large-size cell may indicate different reaction mechanisms across the large-size cell along the gas flow direction. Therefore, the studies of the reaction mechanism on large-size cells were the focus of this paper.

1.3 Reaction mechanism reflected on large-size cells

Limited work focusing on the large-size cells indicated that H_2O electrolysis dominated the performance of co-electrolysis and only in the case of higher CO_2 concentration in the inlet mixture and higher reactant conversion rate (higher applied current) could CO_2 electrolysis occur. The relevant experiments and results were summarized in Table 1.

Kim et al.[10] explored the reaction mechanism of co-electrolysis in the case that the concentration of H_2O was equal or higher than that of CO_2. A planar cell with an active area of 25 cm² with a structure of Ni-YSZ|YSZ|GDC (gadolinium-doped ceria, the same as CGO)|LSC (strontium-doped lanthanum cobalt)-GDC was tested. It was found that H_2O was the primary reactant during co-electrolysis and CO_2 contributed to the electrochemical reaction through replenishing steam through RWGS reaction. Stoots et al.[24] from Idaho National Laboratory announced a 50% higher area specific resistance (ASR) of CO_2 electrolysis than that of steam electrolysis according to the testing results of a button cell and a ten-cell stack. Given the almost overlapping polarization characteristics (*j*-*V* curves) of H_2O electrolysis (54.9% H_2O+22.5% H_2) and co-electrolysis (54.9% H_2O+22.6% CO_2+22.5% H_2), they stated that no CO_2 was electrochemically split and CO was produced only by the RWGS reaction. Similar results were obtained by Thaler et al.[25] on a 5-cell stack showing no clear difference in cell performance between H_2O electrolysis (50% H_2O+50% H_2) and co-electrolysis (33.3% CO_2+33.3% H_2O+33.3% H_2). Therefore, they claimed that only H_2O was electrochemically reduced while CO_2 was only converted by RWGS reaction.

Some other studies were also conducted with higher CO_2 concentration in the inlet gas supply. Li et al.[27] who studied the reaction mechanism of co-electrolysis of 45% CO_2+45% H_2O+10% H_2 and 70% CO_2+10% H_2 on a 63 m² Ni-YSZ|YSZ|LSM (lanthanum magnetite doped with strontium)-YSZ planar cell at 800 °C.

They stated that H_2O electrolysis first started and CO_2 electrolysis initiated and parallelly occurred with H_2O electrolysis at high current during co-electrolysis and the occurrence moment of CO_2 electrolysis was affected by the inlet mixture. Similarly, it was indicated by Wu et al.[28] that the current density may affect the amount of CO_2 that was electrochemically reduced on a 60 cm² flat-tube single cell with the structure of Ni-YSZ|YSZ|GDC|LSCF (strontium and iron-doped lanthanum cobalt)-GDC. Similar results were obtained in our group[23] and it was found that composition of inlet mixture significantly affected the occurrence moment of individual CO_2 electrolysis through affecting the concentration of H_2O. For mixture of 50% CO_2+50% H_2, H_2O electrolysis dominated the performance of co-electrolysis during the loading process from open circuit voltage (OCV) to 1.37 V. The contribution of electrochemical CO_2 reduction on the CO production was also calculated experimentally and it was found that 77.2% and 58.0% of CO was acquired from CO_2 direct electrochemical reduction for mixture of 90% CO_2+10% H_2 and 70% CO_2+30% H_2 at 0.2 A·cm⁻² respectively. Earlier, similar results reported by Ebbesen et al.[26] tested the performance of co-electrolysis on a Ni-YSZ based planar cell of 16 cm². Based on the position of ASR of co-electrolysis (25% H_2O+25%CO_2+25%CO) which was in between that of H_2O electrolysis (50% H_2O+25% H_2) and CO_2 electrolysis (50% CO_2+25% CO), it was claimed that the electrochemical reduction of CO_2 and H_2O both occurred during co-electrolysis.

It can be seen that, even different operating conditions were applied and different conclusions were reached, reported literature shared something in common. In the case of higher H_2O concentration than CO_2 concentration in the inlet, H_2O electrolysis dominated during co-electrolysis and barely CO_2 electrolysis was observed[10, 24, 25]. While for mixtures with higher CO_2 concentration than that of H_2O, CO_2 electrolysis was possible to occur[19, 20, 23, 26–28] and the occurrence moment was related with the amount of H_2O injected or produced through RWGS reaction and the reactants conversion rate or the applied current. At a lower applied current, H_2O electrolysis dominated the performance of co-electrolysis. With the increase of applied current, CO_2 electrolysis initiated and gradually dominated. The current where CO_2 electrolysis occurred and dominated was postponed if more H_2O existed. In terms of the exact occurrence moment of CO_2 electrolysis, different cells may differ and the reaction mechanism needs to be further verified on large-size cell level.

Table 1 Performance tests of co-electrolysis and corresponding reaction mechanisms

Cell configuration	Operating condition	Reaction mechanism	Ref.
Ni-YSZ|YSZ|GDC|LSC-GDC 25 cm² planar cell	800 °C 45% H_2O+22.5% CO_2+10% CO; 45% H_2O+22.5% CO_2; 45% H_2O+22.5% CO_2+10% H_2		[10]
Nickel cermet|scandia-stabilized zirconia| strontium-doped manganite 10-cell stack (planar cells, 64 cm²×10)	800 °C 54.9% H_2O+22.6% CO_2+22.5% H_2; 54.9% H_2O+22.5% H_2; 100% CO_2	H_2O electrolysis dominated; CO_2 was reduced mainly through RGWS reaction.	[24]
Ni-YSZ|YSZ|GDC|LSCF 5-cell stack (planar cells, 80 m²×5)	750 °C 33.3% CO_2+33.3% H_2O+33.3% H_2		[25]
Ni-YSZ|YSZ|GDC|LSCF-GDC 100 cm² planar cell	800°C 90% CO_2+10% H_2; 70% CO_2+30% H_2; 50% CO_2+50% H_2	CO_2 electrolysis occurred at a certain conditions and H_2O amount injected or produced from RWGS reaction and the applied current density decided the occurrence and dominance of CO_2 electrolysis.	[23]
Ni-YSZ|YSZ|LSM-YSZ 63 m² planar cell	800 °C 45% CO_2+45% H_2O+10% H_2; and 70% CO_2+10% H_2		[27]
Ni-YSZ|YSZ|LSM-YSZ 16 m² planar cell	850 °C 25% H_2O+25% CO_2+25% CO; 45% H_2O+45% CO_2+10% H_2; 50% H_2O+25% H_2; 50% CO_2+25% CO	Parallel occurrence of H_2O electrolysis and CO_2 electrolysis.	[26]
Ni-YSZ|YSZ|GDC |LSCF 60 cm² flat-tube single cell	750 °C 75% CO_2+25% H_2	Current density may affect the amount of electrochemically reduced CO_2.	[28]

2 Durability of co-electrolysis on large-scale single cells and stacks

2.1 Durability of co-electrolysis on large-scale single cells

For SOEC technology commercialization and large application in energy storage sector and syngas production, durability is one of the essential issues. Just like researches in solid oxide fuel cell (SOFC), the electrode materials, the structure of cells, and the operating conditions of cells were all essential for the durability of SOEC. However, the investigation about the electrode materials and the structure of cells were most conducted on button cell level and the application of classical Ni-based cathode supported cell with LSM-YSZ anode or LSCF-GDC anode together with a buffer layer of GDC in between anode and electrolyte was overwhelming on large-size cells. What was more concerned on the large-size cell level that affecting the durability of SOEC co-electrolysis was the operating conditions such as inlet composition, working mode (galvanostatic mode or potentiostatic mode), the applied current density or voltage, etc.

Most of the studies investigating the durability of co-electrolysis were conducted in galvanostatic mode. Actually, the cell could be also operated in potentiostatic mode. Working at a constant voltage was uniquely significant for SOEC as it did not need external heat supply if it was operated at thermoneutral voltage in which the heat demand of endothermic splitting reaction can be exactly met by the Joule heat. The degradation comparison of large-size cells operating in different modes was illustrated in Ref. [31]. It was found that similar degradation rates of 2%/kh in two modes were found during the last 800 h operation and working in potentiostatic mode was beneficial in a way as some over-potential related degradation can be suppressed. However, a changing resulting current density may lead to a variation in syngas production rate and syngas composition. Thus, in the perspective of acquisition of stable syngas production, operating SOEC in galvanostatic mode was better to be adopted. And, it was not surprising that the dura-

bility of SOEC was highly dependent on the working current[32, 33] and the durability of SOEC co-electrolysis operated at relative lower current ($\leqslant$0.5 A·cm⁻²) on large-size cells was already demonstrated. Stable performance and low degradation and even no degradation was observed during hundreds and even thousands of hours' operation[34–37]. However, SOEC only merited in perspective of engineering significance when operating at high current (> 0.5 A·cm⁻²) because of the need of massive syngas production and lowering cost. Therefore, only SOEC co-electrolysis working at high current was discussed in this paper.

Literature reporting the results of durability tests based on large-size cells was very limited, most of which was reported by the researchers from Technical University of Denmark (DTU). The operating conditions of these planar cells were summarized in Table 2.

In general, it was found from Table 2 that cells with anode of LSM-YSZ were operated at 800 °C or higher temperature while cells with LSCF-CGO anode were operated at 800 °C or lower temperature such as 750 °C, which may be due to the fact of lower performance and higher degradation rate of cells with LSM-YSZ anode at high current compared with that with LSCF-CGO anode[38]. The degradation rate was reported to be on the order of hundreds of mV per thousand hours[32, 33, 38–41]. A lowest degradation rate of only 12 mV (0.9%)/kh was reported during 2700 h operating at 1 A·cm⁻² (reactant conversion rate of 31%) at 800 °C with a mixture of 45% CO_2+45% H_2O+10% H_2 injected[42]. Other studies also reported tens of mV loss per thousand hours and a degradation rate less than 10%/kh during thousands of hours of operation[31, 43]. The main degradation mechanisms such as adsorption of impurities[32], delamination between cathode and electrolyte[39], formation of carbon nanotubes[39], reduction of YSZ in cathode[39, 42], depletion of Ni percolation, contact loss between Ni and YSZ[41] etc., were reported.

Except from that, it can be seen from Table 2 that most of the durability tests of co-electrolysis at high current were conducted with an inlet mixture of equal concentration of H_2O and CO_2 (45%

Table 2   durability tests of SOEC co-electrolysis at high current on large-size cells

Year	Cell configuration	Operating conditions	Durability test	Degradation rate	Ref.
2011	Ni-YSZ|YSZ|LSM-YSZ 16 cm²	850 °C 45% CO_2+45% H_2O+10% H_2	0.25 A·cm⁻² for 382 h 0.5 A·cm⁻² for 206 h 1 A·cm⁻² for 212 h	0.005–0.008 mV/h on the order of 0.1 mV/h	[32]
2012	Ni-YSZ|YSZ|LSM-YSZ 16 cm²	850 °C 45% CO_2+45% H_2O+10% H_2 Conversion rate 60%	1 A·cm⁻² for 932 h 1 A·cm⁻² for 60 h and 1.5 A·cm⁻² for next 932 h	0.216–0.422 mV/h 0.633–1.3 mV/h	[33]
2013	Ni-YSZ|YSZ|LSM-YSZ 16 cm²	850 °C 45% CO_2+45% H_2O+10% H_2 Conversion rate ⩽66.8%	1.5 A·cm⁻² for ~700 h 2 A·cm⁻² for ~300 h 2 A·cm⁻² for ~700 h 2 A·cm⁻² for ~50 h 2.25 A·cm⁻² for < 50 h	~0.2 mV/h ~0.4 mV/h 5 mV/h	[39]
2013	Ni-YSZ|YSZ|LSM-YSZ 16 cm² Ni-YSZ|YSZ|CGO|LSCF-CGO 16 cm²	800 °C 45% CO_2+45% H_2O+10% H_2 Conversion rate 62%	1.5 A·cm⁻² for ~300 h 1.5 A·cm⁻² for ~400 h	500–1500 mV /kh 300–600 mV /kh	[38]
2013	Ni-YSZ|YSZ|CGO|LSCF-CGO 16 cm² Ni-YSZ|YSZ|LSM-YSZ 16 cm²	800 °C 45% CO_2+45% H_2O+10% H_2 Conversion rate 60%–62%	1.5 A·cm⁻² for 584 h 1.5 A·cm⁻² for 526 h 1 A·cm⁻² for 1256 h	414 mV/kh 504 mV/kh 122 mV/kh	[40]
2014	Ni-YSZ|YSZ|CGO|LSC-CGO 16 cm²	800 °C 45% CO_2+45%H_2O+10% H_2 Conversion rate 31%	1 A·cm⁻² for 2700 h	12 mV (0.9%)/kh	[42]
2016	Ni-YSZ|YSZ|LSM-YSZ 16 cm²	800 °C 45% CO_2+45% H_2O+10% H_2 Conversion rate 45%–60%	1.5 A·cm⁻² for 682 h 2 A·cm⁻² for 678 h	275 mV/kh 452 mV/kh	[41]
2018	Ni-YSZ|YSZ|CGO|LSCF-CGO 16 cm²	750 °C 65% H_2O+25% CO_2+10% H_2 Conversion rate: 38.5%–48.7%	0.75 A·cm⁻² for 1000 h 1.2 V for 1000 h	0.13 mV (10%)/kh; 2%/kh in terms of ASR; 1.3 mA/h (14%/kh); 2% /kh in terms of ASR;	[31]
2021	Ni-YSZ|YSZ|CGO|LSCF-CGO 16 cm²	800 °C 45% CO_2+45% H_2O+5% H_2+5% CO Conversion rate 56% 800 °C 45% CO_2+45% H_2O+5% H_2+5% CO Conversion rate 28%; 800 °C 33% CO_2 + 32% H_2O + 18% H_2+17% CO Conversion rate 39%	1 A·cm⁻² for 1030 h 1 A·cm⁻² for 1190 h 1 A·cm⁻² for 1063 h	337–430 mV/kh 20–140 mV/kh 64 mV/kh	[43]

CO_2+45% H_2O+10% H_2), in which case CO_2 electrolysis would occur, while limited research was conducted at the condition of low CO_2 electrolysis occurrence probability (65% H_2O+25% CO_2+ 10% H_2)[31]. Actually, the selection of inlet gas composition and the applied or resulting current may affect the reaction mechanism of co-electrolysis as indicated in Section 1.3. As showed in Table 2, the relative lower degradation rate was achieved apparently at high applied current density of 1 A·cm⁻², but with a conversion rate of no more than 40%[42, 43], in which the performance of co-electrolysis may be dominated by H_2O electrolysis due to the low oxidant conversion rate. Operating in the interval of H_2O electrolysis dominance, CO_2 electrolysis dominance or H_2O and CO_2 parallel electrolysis dominance may affect the durability of SOEC co-electrolysis. However, this kind of effect could not be fully extracted from the currently reported literature. Furthermore, it should be mentioned that one of the selling points of SOEC co-electrolysis was the achievable tunable syngas ratio of H_2/CO in one reactor[44]. From the perspective of carbon neutrality, high CO_2 conversion rate was required. Based on these two demands, the durability of co-electrolysis with higher CO_2 concentration in the inlet, higher CO_2 conversion rate and tunable H_2/CO ratio should be fully demonstrated.

During the durability tests, the phenomenon of fast and severe degradation during the initial hundreds of hours' (⩽ 350 h) operation was repeatedly reported at small or high current density, on LSM-YSZ or LSCF oxygen electrode and galvanostatic or potentiostatic mode[31, 35, 37, 39, 41, 42, 45]. An example was showed in Figure 4 from Ref. [31]. It was clearly showed that the initial degradation was decisive to the performance of cells after long-term operation. The degradation mechanism and the mitigation method should be further explored to improve the lifetime of cells.

2.2   Durability of co-electrolysis on stacks

The SOEC stack is the core component of an SOEC system and its lifetime is decisive on the durability of a system. Studies concerning the durability tests on stack level were limited and summarized in Table 3.

What should be mentioned was that the durability of stack was hard to characterized as the performance of the cell fluctuated due to the apparent reactivation or degradation induced by the temperature drift, contact problem, unstable gas supply, etc.[17, 46, 47]. The degradation rates reported on stacks were comparable and even lower than that reported on single cells. The problems such as contact problem of the bottom unit[17, 46, 47], depletion of Ni at the
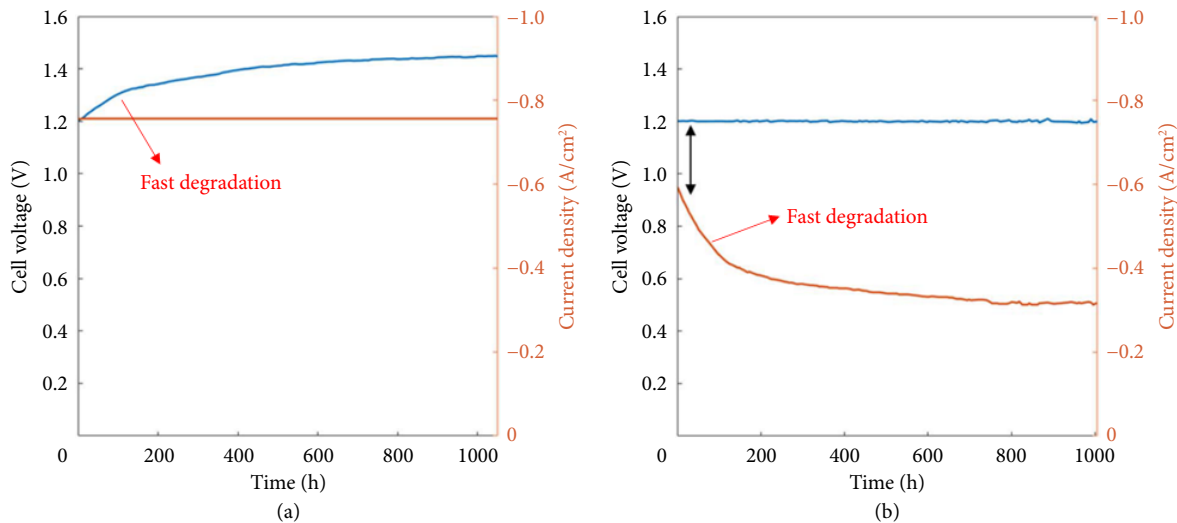
Fig. 4 Current density and cell voltage evolution as function of time during the (a) galvanostatic operation on a 16 cm^2 planar cell, (b) potentiostatic testing on another 16 cm^2 planar cell with 65% H$_2$O+25% CO$_2$+10% H$_2$ supplied to the fuel electrode at 750 °C. Reprinted with permission from Ref. [31], © 2018 Author(s).

Table 3 Durability tests of co-electrolysis on stacks

Year	Stack configuration	Operating conditions	Durability test	Degradation rate	Ref.
2011	10-cell stack (92.2 cm^2 × 10) Ni-YSZ|YSZ|LSM-YSZ	850 °C 45% CO$_2$+45% H$_2$O+10% H$_2$ Conversion rate 60%	0.5 A cm^{-2} for the first 800 h	9 mV/kh	[46]
			0.75 A cm^{-2} for another 350 h	−100 mV/kh	
2013	10-cell stack with three types of cells (92.2 cm^2 × 10)	A: Ni-YSZ|YSZ| LSM-YSZ B: Ni-YSZ|YSZ| LSM-YSZ C: Ni-YSZ|YSZ| CGO|LSCF-CGO	800 °C 45% CO$_2$+45%H$_2$O+10% H$_2$ conversion rate 60%	0.5 A cm^{-2} for 70 h and then 0.75 A cm^{-2} for 1000 h	quite noisy picture; voltage fluctuation 200-400 mV; Activation after durability test Degraded after durability test	[17]
2016	8-cell stack (87.7 cm^2 × 8) Ni-YSZ|YSZ|CGO|LSCF-CGO	45% CO$_2$+45%H$_2$O+10% H$_2$ conversion rate 39%	700 °C, 0.25 A cm^{-2} for 120 h	1.3 V→1.6 V; severe degradation	[47]
			750 °C, 0.25 A cm^{-2} for 2200 h	large activation	
			750 °C, 0.5 A cm^{-2} for 3700 h	17 mV (1.4%)/kh	
2019	4-cell stack (80 cm^2 × 4) Ni-YSZ|YSZ|CGO|LSCF	800 °C 55% H$_2$O+35% CO$_2$+10% H$_2$ and 60% H$_2$O+30% CO$_2$+10% H$_2$ Conversion rate of 70%	≥0.5 A cm^{-2} for around 1700 h	22 mV (~2%)/kh	[48]
2020	2-cell stack (100 cm^2 × 2) Ni-YSZ|YSZ|CGO|LSCF	800 °C 55% H$_2$O+35% CO$_2$+10% H$_2$ Conversion rate of 59%	0.5 A cm^{-2} started at 1550 h and lasted for 1250 h	40–50 mV/kh	[49]
2021	4-cell stack (80 cm^2 × 4) Ni-YSZ|YSZ|CGO|LSCF	800 °C 60% H$_2$O+30% CO$_2$+10% H$_2$ Conversion rate ≥ 70%	0.7 A cm^{-2} for around 800 h	Large degradation rate.	[50]

interface of electrolyte and cathode and possible poisoning of cathode were reported[48, 50]. The feasibility of SOEC on stack level was fully demonstrated.

Similar to the investigation on single cells, mixture with $p_{H_2O} \geqslant p_{CO_2}$ was most adopted while the applied current was limited to be equal or less than 0.75 A·cm^{-2}. Even though the applied current seemed to be lower than the applied current reported in Section 2.1 on single cells, the achieved oxidant conversion rate of 60% and even 70% was high enough and comparable with that on single cells. The reaction mechanism of these stack units whether they were operated in H$_2$O electrolysis dominated interval or CO$_2$ electrolysis dominated interval or H$_2$O and CO$_2$ electrolysis dominated interval at these conditions was hard to distinguish. However, it was indicated that there may be a reaction mechanism change from water splitting to direct CO$_2$ electrolysis during co-electrolysis of 60% H$_2$O+30% CO$_2$+10% H$_2$ when the conversion rate was

higher than 60%[48, 50]. Furthermore, a clear dependency of the degradation rate of co-electrolysis on the conversion rate was observed in Ref. [50]. The degradation rate reduced significantly when the conversion rate was reduced from 70% to 60% but did not change as much with further reduction in conversion rate. However, it was hard to determine whether this kind of degradation was due to the shortage of convertible oxidant shortage due to high conversion rate or the reaction mechanism change. Due to the need of massive conversion of CO$_2$, more future tests should be conducted on stack level with higher inlet CO$_2$ concentration and at higher applied current and the relation between degradation rate and reaction mechanism of co-electrolysis should be figured out. Besides, it was worth noting that the fast and severe degradation during initial operation was also observed when single cells were integrated into a stack[46]. The relevant mitigation strategies should be also further investigated.
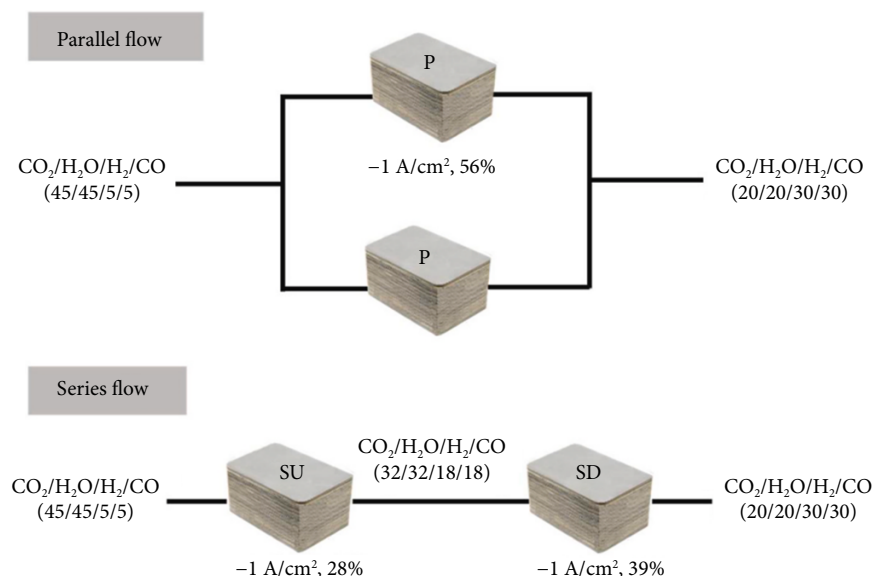
Fig. 5 Schematic illustration of the two operation scenarios from Ref. [43], where Cell P was operated to simulate the parallel flow and Cells SU and SD were operated to simulate series flow upstream (SU) and downstream (SD), respectively. Note: –1 A·cm^{-2} here means operating in SOEC mode at 1 A·cm^{-2}. Reprinted with permission from Ref. [43], © 2021 Author(s).

Different from single cells, stacks encountered the issue of integration, including the integration between single cells and connection of multiple stacks. The reported bottom cell contact problem emphasized the significance of the demand to improve the design of the stack. Meanwhile, the connection of multiple stacks in terms of gas flow was investigated and mimicked in Ref. [43] (summarized in Table 2). Two cells (Cell SU and Cell SD) simulated the upstream and downstream stack in a serial gas-flow arrangement respectively while a third cell (Cell P) was applied to mimic the behavior of stacks with parallel gas flow arrangement as showed in Figure 5 from Ref. [43].

All of the three cells were operated at 1 A·cm^{-2} and the overall conversion rate of two arrangement was equal as 56%. The degradation rate for the cell P was 337 mV/kh while that of Cell SU and Cell SD showed a much lower degradation rate of 140 mV/kh. The great unevenness of the current density distribution and thus great over-potential gradient along the gas flow direction resulted in a higher overpotential for Cell P at the inlet and finally a severer degradation compared with Cell SU and Cell SD. It was found that the serial gas-flow arrangement was preferred as stacks with this kind of arrangement showed reduced degradation and lower power consumption.

2.3 Dynamic operation of SOEC co-electrolysis

The most envisaged application scenario of SOEC system is to combine with renewable energy and use the excess electricity of renewable energy to produce syngas. The compatibility of SOEC system with the intermittent renewable energy is a critical issue. The feasibility of dynamic operation of SOEC H_2O electrolysis was already demonstrated on single cells and stacks[51–53]. Some other studies also tried to demonstrate the feasibility of SOEC co-electrolysis system integration with fluctuating power supply. In 2019, researches from Forschungszentrum Jülich[54] reported the response of a stack numbered F1004-95 to the grid with varying loading profile. The load profile was developed from a power supply scenario for year 2050 for residual application. Different demand at different moment of the day required an output current density

varying from 0.33 A·cm^{-2} to 0.71 A·cm^{-2} and no degradation was observed after a 40-cycle accelerated test. Later in 2021, they tested two 4-cell stacks under load cycles with static gas supply as showed in Figure 6(a) and dynamic gas supply as showed in Figure 6(b) for over 1300 h[50]. Stable syngas production rate and H_2/CO ratio around 2 were acquired during the dynamic operation and only a short-term disturbance of H_2/CO ratio and minor stack overshoots were observed during transitions. The disturbance lasted for a short time and quickly got stabilized. The feeding gas adjustment and re-stabilization involved in a 25% power switchover can be done within 2 minutes. The capability of SOEC co-electrolysis to follow the grid-related requirement profiles of secondary control power and minute reserves was fully demonstrated. Earlier in 2020, researchers from DTU[55] tested the durability of a 6-cell stack with dynamic load using a 24 h wind profile from Bornholm in Denmark for 1000 h as showed in Figures 6(d) and 6(e). Each unit showed a structure of Ni-YSZ|YSZ|CGO|LSC-CGO and an active area of 80 cm^2. During the dynamic test, the constant flow of 65% H_2O+25% CO_2+10% H_2 was supplied and the maximum loading current was no more than 0.5 A·cm^{-2}. A syngas ratio H_2/CO of 3.7 was acquired and an accumulated syngas of 258 m^3 at 25 °C was achieved. The overall degradation rate was only around 0.8%/1000 h. Besides, in 2022, Wu et al.[28] mimicked the response of co-electrolysis (75% CO_2+25% H_2) towards intermittent renewable energy by imposing a pulsed current on a flat-tube single cell of 60 cm^2 with structure of Ni-YSZ|YSZ|GDC|LSCF-GDC. 100 cycles with each cycle lasting for 8 h and pulsed current varying from 0.1 A·cm^{-2} to 0.3 A·cm^{-2} were performed and a degradation rate of 0.041% per cycle was reported when a higher current density of 0.3 A·cm^{-2} was applied. Meanwhile, a lifetime of 500 cycles was estimated. These results preliminarily demonstrated the great compatibility of SOEC co-electrolysis with fluctuating power supply from renewable energy and the great application potential in the field of energy storage and negative emission.

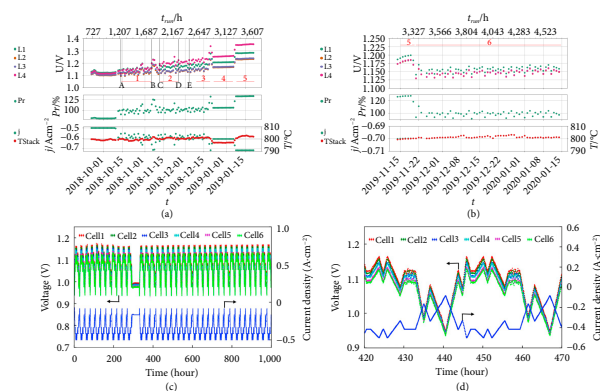What should be noted was that the choosing of operating con-

Fig. 6 Daily averaged layer voltages (top), relative power (center), current density, and stack temperature (bottom) for **(a)** stack A with static gas supply and **(b)** stack B with dynamic gas supply[50]; **(c)** Voltage evolution of a 6-cell stack tested under dynamic load conditions[55]; **(d)** a zoom image into 50 h of the 6-cell stack from DTU for better visibility of the response of each unit[55]. Reprinted with permission from Ref. [55], © 2022 Elsevier.

ditions should be well thought out. During the realistic dynamic operation, the production rate and composition of syngas would be affected when only current density was changed and gas supply was constant. Also, the maximum applied current or the highest conversion rate should be also considered as it may cause great degradation on the stack. Besides, when the stack was operated under dynamic gas supply, the quick adjustment of feeding gas especially the supply of steam should give a response as quick as possible, which raised a higher demand on the equipment. In summary, the operating conditions of a stack or system should be optimized considering the realistic requirements such as obtaining a stable syngas yield and ratio as well as less degraded stack performance and the limits of relevant equipment.

3　Summary and outlook

SOEC was a promising technology in the field of energy storage and negative carbon emission, but there is still a long way to go in terms of its future massive application and commercialization. The coupling of RWGS reaction and the electrochemical reactions make the reaction mechanism of co-electrolysis complicated. It was found that explorations on button cells could not reflect the reaction mechanism on large-size cell as it could not mimic the gas phase composition and current density gradient along the flow direction due to a low reactant conversion rate and an electrode surface-perpendicular gas supply path. On large-size cells, H_2O electrolysis always dominated the performance of co-electrolysis and CO_2 electrolysis occurred and even dominated only in the case of higher CO_2 concentration than H_2O in the inlet and with high conversion rate (high applied current). Durability was also one of the essential issues limiting the commercialization of SOEC co-electrolysis. The durability of co-electrolysis at high applied current (> 0.5 A·cm⁻²) was summarized and analyzed. The selection of operating conditions and the corresponding reaction mechanism at these operating conditions may play a role in the durability of single cells and stacks. Besides, the phenomenon of fast and severe degradation was emphasized on single cells and stack units. Finally, the feasibility of dynamic operation of SOEC co-electrolysis and its application potential in the field of energy storage and negative carbon emission was illustrated.

Except for that, some issues needed to be considered and mitigated seriously.

(1)　Most of the reaction mechanism studies were conducted on button cells and the reaction mechanism of co-electrolysis on large-size cells should be further verified.

(2)　The relation between the reaction mechanism behind the chosen operating conditions and the durability of single cells and stacks should be figured out.

(3)　The degradation mechanism and the mitigation of fast initial degradation should be explored.

(4)　Further researches on stack design and operating strategy under dynamic load and towards sudden problems should be developed.

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Article history

Additional information

Declaration of competing interest

The authors have no competing interests to declare that are relevant to the content of this article.

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