Effect of Simulated Coal-Derived Gas Composition on H₂S Poisoning Behavior Evaluated Using a Disaggregation Scheme

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H₂S poisoning is an important issue for solid oxide fuel cells (SOFCs) operated with syngas. The effect of simulated coal-derived gas composition on H₂S poisoning behavior was evaluated using a disaggregation scheme where the influence of H_2 content was determined separately using a typical anode-supported SOFC operated with a N_2/H_2 mixture gas, while the effect of other compositions (CO, CO_2, and H_2O) was investigated with simulated coal-derived gas having constant H_2 and CO flow rates balanced by a H_2/N_2 mixture gas (83% H_2 and 17% N_2). The results indicated that the extent of H₂S poisoning was not pertinent to H_2 content when the cell was tested galvanostatically with a current density of 0.3 A/cm² at 800°C using a N_2/H_2 mixture gas containing 10 ppm H₂S, and the H₂S poisoning impact can be completely removed by switching to sulfur-free gas. The CO, CO_2, and high water vapor content aggravated the H₂S poisoning effect, and the performance was almost irrecoverable when the cell was tested with a 35% H_2–46% CO–16% N_2–3% H_2O mixture gas containing 12.5 ppm H₂S. However, the introduction of 10% CO_2 and an increase in H_2O content from 3 to 10% in the mixture gas can promote the performance recoverability to a larger extent.
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A wide variety of fuels, including coal gas, natural gas, biogas, and gasoline, can be used in a solid oxide fuel cell (SOFC) system even without pre-reforming. However, fuel impurities in coal syngas have been reported to deteriorate cell performance and shorten its life.[1] Because coal gas contains tens to thousands parts per million of H₂S, and because a few parts per million of H₂S in fuel can cause a great drop in cell power output, much attention has been paid to the behavior and mechanisms of H₂S poisoning on SOFC.[2-8] Two different degradation mechanisms of H₂S poisoning on Ni/yttria-stabilized zirconia (YSZ) cermets can be described as a sharp drop in cell performance caused by adsorption of S_x on the surface of a nickel catalyst and a slow persistent degradation due to the formation of Ni_xS_y. Both mechanisms are strongly dependent on operating temperatures.[9,10]

Many investigations have been conducted on the poisoning behavior of H₂S on the SOFC operated on pure H_2 or the mixture gas of H_2 and N_2. Ishikura et al.[2] studied the influence of H₂S concentration from 0.1 to 20 ppm on cell output voltage using H_2/N_2 as fuel when the cell was operated under a constant current density of 200 mA/cm² at 900°C. It was shown that the output voltage dropped to zero as the cell was exposed to 20 ppm H₂S and was not completely recovered after the removal of H₂S. Rasmussen and Hagen[3] investigated the cell voltage drop caused by the addition of 2 to 100 ppm H₂S in H_2 and suggested that 40 ppm H₂S content was the saturation point.

Recently, some studies have been conducted focusing on the poisoning behaviors of H₂S on the SOFC fueled by the coal-derived gas. Trembly et al.[8] reported that the introduction of 200–240 ppm H₂S led to a 10–12.5% degradation of cell performance with a simulated syngas of CO–H_2–N_2–H_2O as fuel. Although the poisoning effect of H₂S is indubitable, there are discrepancies on the poisoning extent of H₂S on SOFC operated with different fuel gases. A 100% degradation was revealed using H_2/N_2 containing 20 ppm H₂S as fuel, while only 10–12.5% degradation can be detected when 200–240 ppm H₂S was added in the syngas fuel composed of CO–H_2–N_2–H_2O. Therefore, the composition of syngas may affect the sulfur poisoning extent on SOFC.

Previous work mainly focused on the poisoning effect of H₂S on cell performance in terms of H₂S concentration, operating temperatures, current density, and cell voltage,[2,3] but for an SOFC system operated with coal-derived gas containing H₂S, it is necessary to systematically diagnose the influence of the composition of coal syngas on H₂S poisoning behaviors. This paper is thus initiated to investigate the effect of the coal syngas composition, including H_2, N_2, CO, CO_2, and H_2O, on the H₂S poisoning behavior for typical anode-supported SOFCs.

Experimental

Typical planar anode-supported 15 × 15 cm SOFCs were manufactured by batch at the Division of Fuel Cell and Energy Technology in Ningbo Institute of Material Technology and Engineering, Chinese Academy of Sciences. A 400 μm thick Ni/8YSZ anode substrate was prepared by tape casting, as were the other cell layers, including a 10 μm thick anode functional layer and a 10 μm thick 8YSZ electrolyte layer. The cathode of the cell was a double $La_{0.8}Sr_{0.2}MnO_3$ (LSM) layer sprayed on the anode substrate before cosintering. The overall cell dimensions for the test were 5 × 5.8 cm with an active area of 4 × 4 cm.

All of the cells were tested in an alumina testing house. Platinum and nickel foil were employed as current collectors at cathode and anode sides, respectively. Silver mesh sandwiched LSM was utilized for gas distribution at the cathode side where silver mesh was aimed to decrease the contact resistance between the cathode and the current collector.[11] For the anode side, two layers of nickel foam were applied for gas distribution and current collection.

Ceramic glass mainly consisting of silicon dioxide was used to achieve good sealing, and an extra 5.5 kg weight was applied to enhance the sealing effect. The cells were then placed into a furnace and heated in N_2 to 850°C with a heating rate of ∼1°C/min to make the sealant intenerate. The reduction of the NiO–YSZ was performed under pure hydrogen with a flow rate of 0.3 standard liter per minute (SLM) for more than 5 h to ensure complete reduction of the anode. The furnace temperature was then decreased to 800°C, which was the testing temperature. The demonstration of the testing facility is shown in Fig. 1.

After the anode was completely reduced, the air was used as a cathode oxidant with a flow rate of 2 SLM, while pure hydrogen was introduced into the anode side as fuel gas with a flow rate of 0.5 SLM. The cell was discharged galvanostatically at 0.3 A/cm² for at least 1 h to ensure sharp electrode activation.[12,13] The current–voltage (*I-V*) curves were recorded at different mixing ratios of H_2/N_2 as anode gas (H_2 content from 27 to 53%). A 10 ppm H₂S balanced by N_2 was subsequently injected into the cell system fueled with the above H_2/N_2 mixture gas for 60 min and then removed. The degradation in cell performance was recorded in terms of voltage drop at a constant discharging current density of 0.3 A/cm². The electrochemical impedance spectra (EIS) were ob-
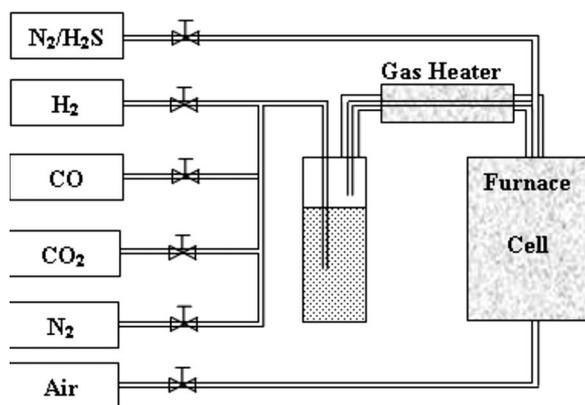
Figure 1. Illustration of the testing facility.

Figure 2. *I-V* curves measured using different ratios of H_2/N_2 at 800°C.

tained at open-circuit voltage (OCV) by four-electrode configuration using an electrochemical workstation (IM6ex, ZAHNER) with a scanning frequency range from 0.1 to 2 MHz before the 10 ppm H_2S injection and 30 min after H_2S removal, respectively. The impedance spectra in galvanostatic mode were also recorded at a current density of 0.3 A/cm² at 800°C for further inspection.

In exploring the effect of the compositions (CO, CO_2, and H_2O) in simulated coal-derived gas on H_2S poisoning behavior, a H_2/N_2 mixture gas (83% H_2 and 17% N_2) was used as the initial base fuel, and CO, H_2O, and CO_2 were added into the base fuel to form mixture gases of 35% H_2–46% CO–16% N_2–3% H_2O, 31% H_2–42% CO–12% CO_2–12% N_2–3% H_2O, and 29% H_2–38% CO–10% CO_2–13% N_2–10% H_2O, where H_2 and CO flows were set as 0.26 and 0.265 SLM, respectively, throughout the experiments for all the mixture gases. The cell was tested under these mixture gases with the injection of 12.5 ppm H_2S for 180 min at a current density of 0.25 A/cm². Impedance spectra were recorded at OCV after removing H_2S for 10 and 660 min, respectively. For comparison, the impedance spectra were also recorded before poisoning. The durability of the cell operated with a simulated coal-derived gas of 29% H_2–38% CO–10% CO_2–13% N_2–10% H_2O was also investigated up to 10 h at a current density of 0.25 A/cm².

Results and Discussion

Investigations were conducted to explore the influence of each composition in simulated coal-derived gas containing H_2, CO, CO_2, H_2O, and N_2 on H_2S poisoning behavior. The H_2 content in H_2/N_2 mixture gas was thus evaluated first, and the effect of the addition of CO, CO_2, and H_2O was subsequently determined.

Effect of H_2 content on H_2S poisoning behavior.— Figure 2 shows *I-V* curves of a single cell fueled with N_2/H_2 mixture gas after the cell is completely reduced for 5 h. It can be seen that the performance of the cell operated at 800°C is moderate. The maximum power output (P_{max}) increases from 287 to 385 mW/cm² with the increase H_2 content from 27% (0.4 SLM) to 53% (0.8 SLM), while the flow rate for N_2 remains constant. The OCV for each composition of H_2/N_2 is almost the same, reaching a potential value of about 1.1 V.

Figure 3a shows the degradation in cell voltage by exposure to 10 ppm H_2S when the cell is fueled with various ratios of H_2/N_2 at a current density of 0.3 A/cm². The cell voltage drops about 3.2–4.2% from the initial voltages of 761–780 mV by 10 ppm H_2S injection for 60 min. and after switching to sulfur-free gas, the cell voltage can almost be fully recovered within 30 min. The output voltage exhibits a small abrupt drop once H_2S is introduced for all five gas compositions and then gradually reaches a stable value after 35 min. When stopping H_2S flow, a small abrupt recovery is observed, and the voltage then immediately returns to its initial value within 25 min. Figure 3b shows the EIS measured at OCV before
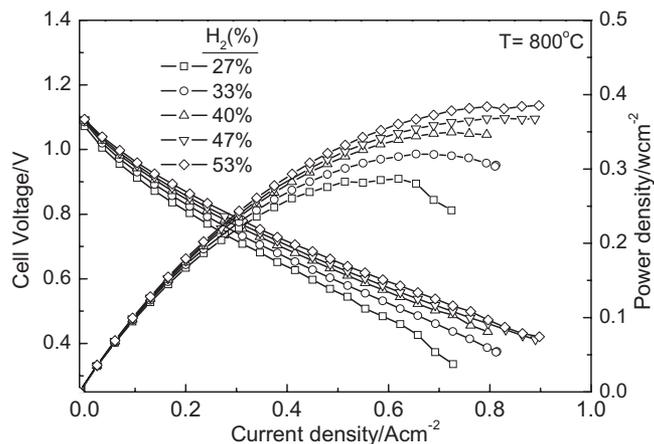
H_2S introduction and after H_2S removal for 30 min. The results obtained before H_2S injection show that the polarization resistance (R_p) increases with the increase in N_2 content, which results in relatively low cell performance. After removing H_2S injection for 30 min, R_p can almost go back to its original value, which agrees with the results from Fig. 4a. Comparing the results before poisoning with the results after stopping H_2S injection for 30 min, no legible deviation was detected for all five H_2 content compositions, indicat-

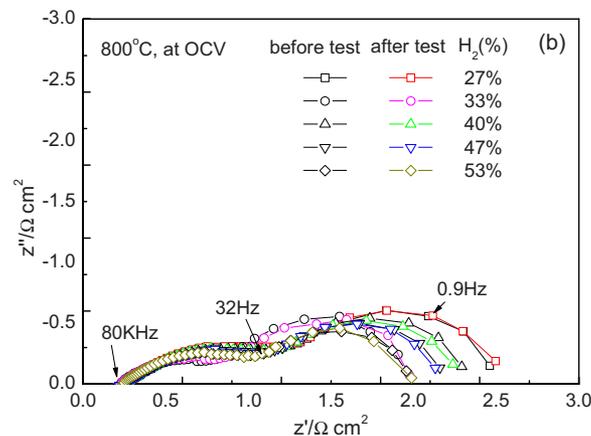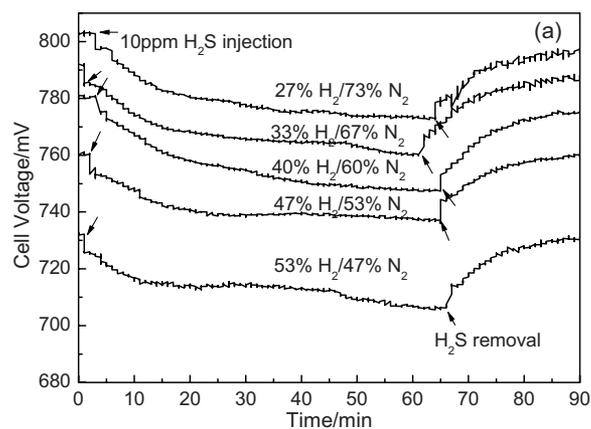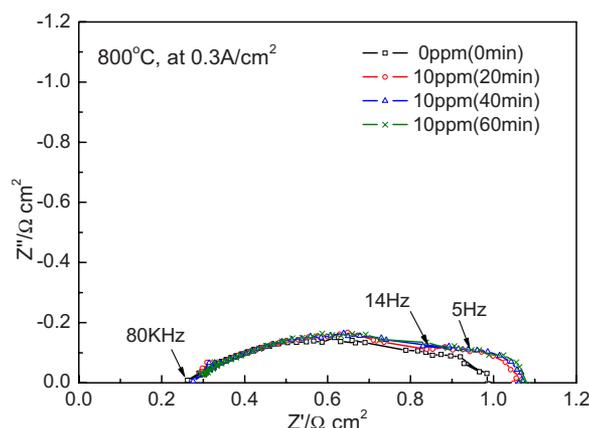**Figure 3.** (Color online) (a) The variation in output voltage for the single cell operated with H_2/N_2 mixture gas at a current density of 0.3 A/cm² and a temperature of 800°C. (b) Impedance spectra at OCV before 10 ppm H_2S injection and removal for 30 min.

Figure 4. (Color online) Impedance spectra curves for the cell fueled with 83% N_2/17% H_2 containing 12.5 ppm H_2S obtained in a galvanostatic mode after H_2S injection for 0, 20, 40, and 60 min, respectively.

ing the recoverability of the H_2S-poisoned cell by switching to sulfur-free gas as reported.[2] The results of impedance spectra show that the R_p increases after injecting H_2S, while R_s remains constant, suggesting that introducing H_2S to fuel can increase the difficulty in some electrochemical processes occurring in the anode side.

The cell voltage drop caused by 10 ppm H_2S is listed in Table I where the cell is operated with H_2/N_2 mixture gases of different ratios. The largest drop in cell voltage is $\sim$4.2% from 780 mV and occurs when the cell is operated with 40% H_2/60% N_2, while the smallest degradation of 3.2% from 761 mV is obtained with the application of 33% H_2/67% N_2. Decreasing H_2 content in the fuel gas cannot decrease the H_2S poisoning effect. This is different from the results reported by Lohsoontorn et al.[4] where polarization resistance and bulk resistance increase with the decrease in H_2 content from 97 to 9.7%. But nevertheless, no evident difference in voltage drop is observed in this study when decreasing H_2 content. Sulfur compound formed at the anode side may be immediately reduced back to sulfur hydrogen in a H_2S free fuel environment at least by thermodynamic consideration. This reduction process is dependent on p_{H_2O}/p_{H_2}.[14] The results in this paper indicate that a 27% H_2 content in N_2/H_2 mixture fuel gas is sufficient to supply a reducing atmosphere to reduce the H_2S poisoning effect. Moreover, the initial voltage (U_0) increases with increasing H_2 content and has no significant influence on the H_2S poisoning effect, which may also be ascribed to the saturation of H_2 content in this study.

To explore the mechanism of the H_2S poisoning cell, impedance spectra were monitored and recorded, as shown in Fig. 4, where the cell was fueled with 33% H_2–67% N_2 at 0.3 A/cm² after injecting H_2S for 0, 20, 40, and 60 min. As shown in Fig. 4, R_p increases rapidly once 10 ppm H_2S is introduced within 20 min and then gradually reaches an equilibrium value until exposure to H_2S for 60 min. This agrees with the results of the electrical properties, as shown in Fig. 3a. The difference in impedance spectra exists in a frequency zone ranging from 0.1 to 5 Hz (14 Hz), which is reported
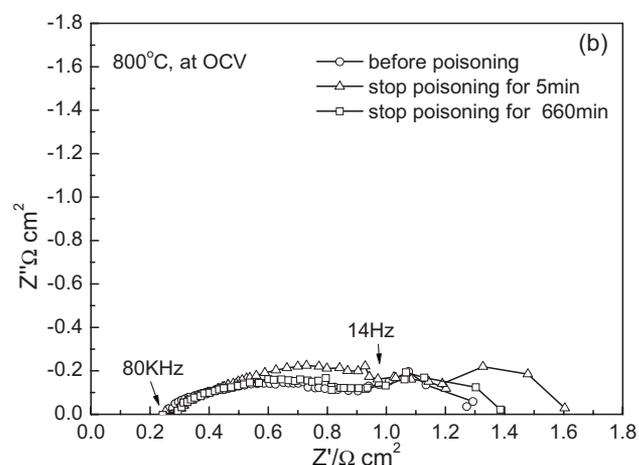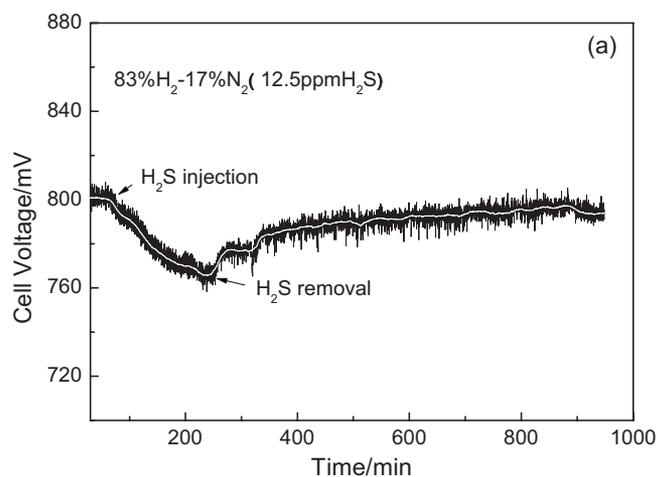
Figure 5. (a) Effect of CO addition to the mixture gas on 12.5 ppm H_2S poisoning behavior and (b) impedance spectra at OCV before and after poisoning.

to involve diffusion and gas conversion at the anode.[15] This indicates that the presence of H_2S can block gas diffusion and bring hindrance in gas conversion at the anode side, thereby leading to an immediate drop in cell performance with the injection of H_2S. However, the microstructural inspection using scanning electron microscope shows that the anode microstructure after poisoning tests is almost the same as the anode without injection of H_2S even for the cell exhibiting highest degradation by H_2S poisoning. The lack of microstructural evidence may be due to short-term poisoning and the small amount of H_2S (10 ppm).[10]

Effect of other compositions of simulated coal-derived syngas.— Figure 5a shows the variation in cell voltage caused by exposure to 12.5 ppm H_2S with a constant discharging current den-

Table I. The drop extent of cell voltage caused by 10 ppm H_2S when operated with different ratios of N_2–H_2 mixture gas at a current density of 0.3 A/cm² and a temperature of 800°C.

|	27% H_2/73% N_2	33% H_2/67% N_2	40% H_2/60% N_2	47% H_2/53% N_2	53% H_2/47% N_2
U_0/mV	732	761	780	792	803
U_{end}/mV	705	737	747	760	772
ΔU (%)	−3.7%	−3.2%	−4.2%	−4.0%	−3.9%
U_r [a] (%)	100%	100%	99.6%	99.4%	99.3%

[a] U_r denotes the recovery percentage of the voltage.
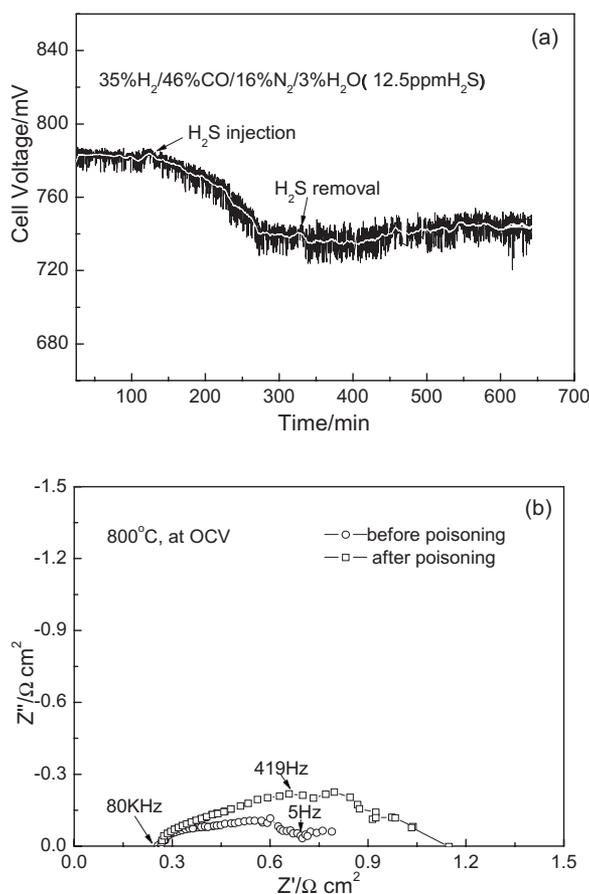
Figure 6. (a) Effect of the CO_2 addition to the mixture gas on 12.5 ppm H_2S poisoning behavior and (b) impedance spectra at OCV before and after poisoning.

Figure 7. (a) Effect of increasing H_2O content on 12.5 ppm H_2S poisoning behavior and (b) impedance spectra at OCV before and after poisoning.

sity of 0.25 A/cm² when the cell is fueled by 83% H_2 and 17% N_2. The voltage drop is 35 mV, equivalent to 4.4% of the initial voltage 805 mV. However, no abrupt drop is observed with the addition of H_2S, which is different from the poisoning behavior discussed above. The voltage tends to continue decreasing without reaching a stable value within 3 h, and the total degradation in the cell is more serious by comparison with the results in Fig. 3a. This may be attributed to the change of silver mesh sandwiched nickel foil to nickel foam as current collector at the anode side because other testing conditions in this test are identical to the previous tests. However, the mechanism for this difference in degradation tendency is not clear. One explanation may be that using nickel foam as the anode for current collecting can suppress a sharp drop in cell performance, thereby demonstrating a persistent gradual degradation. In addition, the voltage is almost completely recovered as soon as sulfur-free gas is flowed in, giving evidence of less poisoning impact with the use of nickel foam. The results derived from impedance spectra are given in Fig. 5b where polarization resistance continues to decrease after stopping H_2S flow for 5–660 min, and the deviation of frequency range is below 14 Hz, indicating that the H_2S poisoning leads to difficulties in some electrochemical processes in the anode; these difficulties can be slowly eliminated once the H_2S is removed.

Figure 6a displays the influence of the introduction of CO into fuel (31% H_2 and 16% N_2) on the H_2S deteriorating effect. The tendency of the initial voltage drop is the same as the cell operating with 83% H_2 and 17% N_2. The degradation percentage reaches 5%, ~45 mV, slightly higher than that using H_2/N_2 as fuel gas (4.4%). However, after removal of H_2S for 300 min, only little recovery is achieved. This indicates that the poisoning for the cell tested in
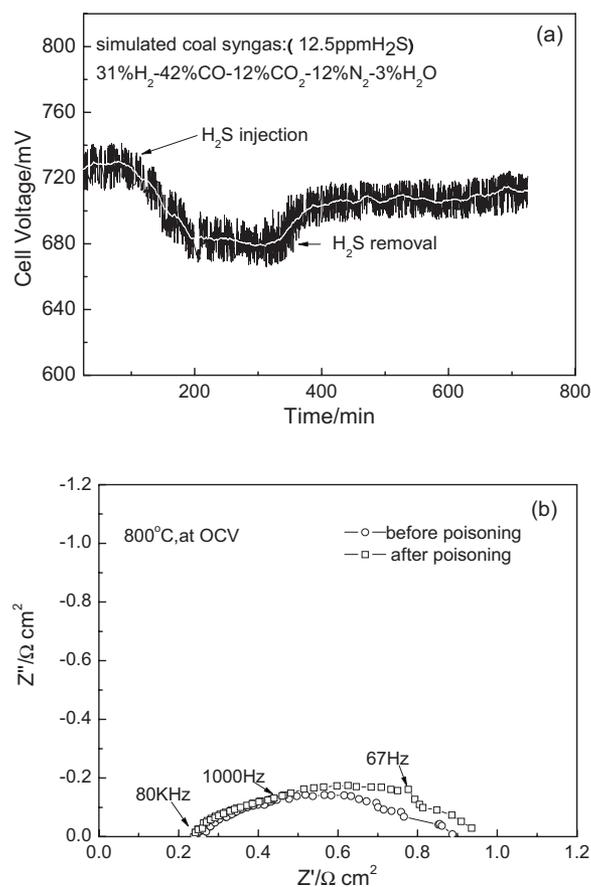
H_2–CO–N_2 cannot be completely removed after stopping H_2S flow, which is further confirmed in Fig. 6b by the results of impedance spectra measured before H_2S poisoning and after stopping H_2S flow for 300 min. The second and third arcs in the impedance spectra increase greatly, representing that the charge transfer between Ni and YSZ is also detrimentally affected.[15,16] This may be attributed to the adsorption of S on the surface of Ni or the formation of Ni_xS_y.[9,10,17]
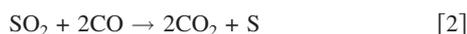
The H_2S poisoning behavior after the addition of 12% CO_2 into the above mixture gas (H_2–CO–N_2) is shown in Fig. 7a. The cell voltage also exhibits a gradual degradation mode, which is identical to the above two tests. However, the degradation percentage increases to nearly 6.9% at ~53 mV, about 1.9% higher than that of the cell operated with H_2–CO–N_2. The voltage is detected to be recovered to about 97% of the initial voltage value after H_2S removal. This indicates that the addition of CO_2 can be beneficial to mitigate the degradation caused by H_2S poisoning. Figure 7b shows the impedance spectra curves obtained before H_2S poisoning and after stopping the H_2S flow when the single cell is fueled with the above H_2–CO–N_2–CO_2 mixture gas. As shown in Fig. 7b, the incomplete recovery of single cell performance is further proved by impedance spectra. The polarization resistance slightly increases after poisoning, whereas the increasing extent is somewhat smaller by comparison with the value obtained from Fig. 6b.

As shown in Eq. 1, H_2S can be electrochemically oxidized to SO_2 by oxygen.[17] Because SO_2 may be present at the cell anode, it would react with CO addition to form CO_2 and sulfur (Eq. 2) resulting in degradation to a larger extent and irrecoverable performance drop

$$H_2S + O_2 \rightarrow SO_2 + H_2O \qquad [1]$$
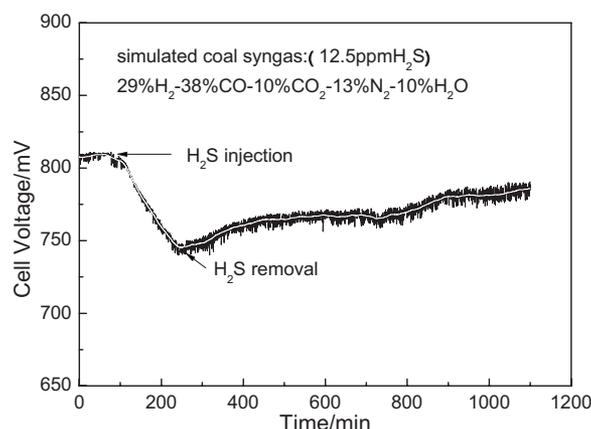
Figure 8. Voltage vs operation time for the single cell operated with a 29% H_2–38% CO–10% CO_2–13% N_2–10% H_2O simulated coal gas containing 12.5 ppm H_2S.

Figure 9. Cell voltage vs operation time for the single cell operated at a current density of 0.25 A/cm^2 and 800°C using a 29% H_2–38% CO–10% CO_2–13% N_2–10% H_2O simulated syngas without H_2S.

$$SO_2 + 2CO \rightarrow 2CO_2 + S \qquad [2]$$

In the mixture gas, a reversible chemical reaction involving CO_2 can be described as Eq. 3 at the operation temperatures of SOFC.[18,19] The addition of CO_2

$$CO_2 + H_2 \leftrightarrow CO + H_2O \qquad [3]$$

can actually increase the CO and water content in fuel gas, leading to a twofold effect on the testing result: (*i*) Enhancement of CO leads to more deterioration in cell performance, as explained by Eq. 2 and (*ii*) water product may convert S_n to H_2S and SO_2, as shown in Eq. 4.[17] The production of H_2S and SO_2

$$H_2S + \tfrac{1}{2}SO_2 \leftrightarrow 2nS_n + H_2O \quad (n \sim 8) \qquad [4]$$

may be partially blown out from the anode gas chamber or electrochemical active zone and thus lead to a partial performance recovery.

To investigate the effect of water content in coal-derived gas on the H_2S poisoning behavior, the vapor content is increased from 3 to 10%,^{1} and thus the fuel composition is determined as 29% H_2–38% CO–10% CO_2–13% N_2–10% H_2O. Figure 8 shows the variation in voltage with time for the cell fueled with 29% H_2–38% CO–10% CO_2–13% N_2–10% H_2O containing 12.5 ppm H_2S. The output voltage for the cell operated with the fuel containing H_2S is degraded to 7.8% at ~63 mV, 0.9% higher than that of the cell operated with 31% H_2 – 42% CO – 12% CO_2 – 2% N_2 – 3% H_2O in Fig. 7a where the fuel has similar components except for the amount of water vapor. After H_2S removal for 800 min, the output voltage recovers to about 97% of the initial value and becomes stable thereafter. It seems that enhancing water content results in a more serious voltage drop, but it is also more helpful to the performance recovery of the single cell fueled with coal-derived gas. As interpreted by Eq. 4, water may hold back the formation of sulfur by the reaction of H_2S and SO_2, thereby permitting a larger performance recovery. At the same time, sulfur can also be produced by H_2S decomposition (Eq. 5)

$$H_2S \rightarrow \tfrac{1}{2}S_2 + H_2 \qquad [5]$$

so that the adsorption of S_2 on the Ni surface may result in an immediate drop in cell voltage. If more S adsorption occurs directly and immediately on the nickel catalyst surface, the S may react with Ni to form Ni_xS_y,[10] leading to a larger performance recovery. The results in Fig. 8 are thus compared with the variation in voltage with operation time for the single cell using the same fuel, 29% H_2–38% CO–10% CO_2–13% N_2–10% H_2O, without H_2S at the same current density, as shown in Fig. 9. Apparently, the cell not exposed to H_2S is operated steadily with the above simulated
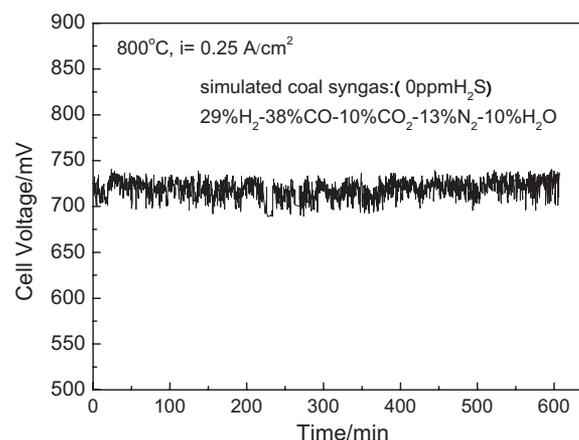
syngas for 600 min without any degradation at 800°C, suggesting that only H_2S causes the cell performance variation.

Conclusions

Investigations have been conducted on the effect of H_2 content in H_2/N_2 mixture gas and the effect of other compositions in simulated coal-derived gas on H_2S poisoning behavior. Increasing the H_2 content from 27 to 53% has no influence on the H_2S poisoning effect. The degradation impact caused by exposure to H_2S is completely removed after H_2S removal when the cell is fueled with H_2/N_2. When the cell is operated with simulated coal syngas, the addition of CO, CO_2, and H_2O aggravates the H_2S poisoning effect; however, increasing the CO_2 and H_2O content may be helpful for performance recovery.

References

1. F. N. Cayan, M. J. Zhia, S. R. Pakalapatia, I. Celika, N. Q. Wua, and R. Gemmenb, *J. Power Sources*, **185**, 595 (2008).
2. A. Ishikura, S. Sakuno, N. Komiyama, H. Sasatsu, N. Masuyama, H. Itoh, and K. Yasumoto, *ECS Trans.*, **7**(1), 845 (2007).
3. J. F. B. Rasmussen and A. Hagen, *J. Power Sources*, **191**, 534 (2009).
4. P. Lohsoontorn, D. J. L. Brett, and N. P. Brandon, *J. Power Sources*, **183**, 232 (2008).
5. S. W. Zha, Z. Cheng, and M. L. Liu, *J. Electrochem. Soc.*, **154**, B201 (2007).
6. K. Sasaki, K. Susuki, A. Iyoshi, M. Uchimur, N. Imamur, H. Kusaba, Y. Teraok, H. Fuchino, K. Tsujimoto, Y. Uchida, et al., *J. Electrochem. Soc.*, **153**, A2023 (2006).
7. K. Sasaki, S. Adachi, K. Haga, M. Uchikawa, J. Yamamoto, A. Iyoshi, J.-T. Chou, Y. Shiratori, and K. Itoh, *ECS Trans.*, **7**(1), 1675 (2007).
8. J. P. Trembly, A. I. Marquez, T. R. Ohrn, and D. J. Bayless, *J. Power Sources*, **158**, 263 (2006).
9. Z. Cheng and M. L. Liu, *Solid State Ionics*, **178**, 925 (2007).
10. J. Dong, Z. Cheng, S. W. Zha, and M. L. Liu, *J. Power Sources*, **156**, 461 (2006).
11. S. P. Jiang, J. G. Love, and L. Apateanu, *Solid State Ionics*, **160**, 15 (2003).
12. V. A. C. Haanappel, A. Mai, and J. Mertens, *Solid State Ionics*, **177**, 2033 (2006).
13. S. Koch, M. Mogensen, P. V. Hendriksen, N. Dekker, and B. Rietveld, in *Proceedings of the Sixth European Solid Oxide Fuel Cell Forum*, European Fuel Cell

Forum, p. 1000, Lucerne, Switzerland (2004).
14. Z. Cheng, S. W. Zha, and M. L. Liu, *J. Power Sources*, **172**, 688 (2007).
15. R. Barfod, A. Hagen, S. Ramousse, P. V. Hendriksen, and M. Mogensen, *Fuel Cells*, **6**, 141 (2006).
16. R. Barfod, M. Mogensen, T. Klemensø, A. Hagen, Y. L. Liu, and P. Vang Hendriksen, *J. Electrochem. Soc.*, **154**, B371 (2007).

17. A. Lussier, S. Sofie, J. Dvorak, and Y. U. Idzerda, *Int. J. Hydrogen Energy*, **33**, 3945 (2008).
18. J. N. Kuhn, N. Lakshminarayanan, and U. S. Ozkan, *J. Mol. Catal. A: Chem.*, **282**, 9 (2008).
19. F. P. Nagel, T. J. Schildhauer, J. Sfeir, A. Schuler, and S. M. A. Biollaz, *J. Power Sources*, **189**, 1127 (2009).