

Short communication

## Influence of cell voltage and current on sulfur poisoning behavior of solid oxide fuel cells

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### Abstract

The sulfur poisoning behavior of nickel-yttria stabilized zirconia (YSZ) cermet anodes in solid oxide fuel cells (SOFCs) was investigated under both potentiostatic and galvanostatic conditions. While the observed relative drop in cell power output caused by sulfur poisoning decreases as the cell-terminal voltage is lowered potentiostatically (thus more current passing through the cell), it increases as more current is drawn from the cell galvanostatically (thus leading to lower terminal voltage). The apparent contradictory trends in relative performance loss due to sulfur poisoning are explained using a simple equivalent circuit analysis, which was further validated by impedance measurements of cells before and after poisoning by trace amounts of hydrogen sulfide ( $\text{H}_2\text{S}$ ) under different conditions. Results suggest that the relative increase in cell internal resistance caused by sulfur poisoning is smaller when more current is drawn from the cell (or the cell-terminal voltage is lowered) under either potentiostatic or galvanostatic conditions. Thus, the increase in anode polarization resistance, not the drop in cell power output, should be used to describe the degree of sulfur poisoning in order to avoid any confusion.

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**Keywords:** Solid oxide fuel cell (SOFC); Hydrogen sulfide ( $\text{H}_2\text{S}$ ); Potentiostatic mode; Galvanostatic mode

### 1. Introduction

The sulfur poisoning behavior of Ni-based anodes for solid oxide fuel cells (SOFCs) has been investigated extensively [1–24]. Most studies show that the initial sulfur poisoning of SOFC anodes is a very fast process [2,4,8,11,13,15–20]. A few studies also suggest that sulfur poisoning effect may continue to develop for tens to hundreds of hours [2,8,17,18,20]. Most previous studies also indicate that the extent of sulfur poisoning increases as the hydrogen sulfide ( $\text{H}_2\text{S}$ ) concentration increases or the cell temperature decreases [2,8,10,13,15,16,18,20].

However, there appears to be contradictory trends in performance loss due to sulfur poisoning under different testing conditions: constant cell current (or galvanostatic condition) versus constant voltage (or potentiostatic condition). For example, Singhal et al. [2] reported that the relative power output drop caused by exposure to 10 ppm  $\text{H}_2\text{S}$  increased from 10.3% to 15.6% when the cell current density increased from 160 to

250  $\text{mA cm}^{-2}$  at 1000 °C. Similarly, Xia and Birss [14] indicated that the relative cell power output drop caused by 10 ppm  $\text{H}_2\text{S}$  increased from 19% to 56% when the current density increased from 130 to 400  $\text{mA cm}^{-2}$  at 800 °C. Waldbillig et al. also reported that when a hydrogen fuel with 1 ppm  $\text{H}_2\text{S}$  was used, the relative drop in cell power output was 6.5%, 9.8%, and 11.8% for a constant cell current density of 250, 500, and 990  $\text{mA cm}^{-2}$ , respectively at 750 °C [15]. Under potentiostatic conditions, in contrast, a previous study shows that the relative drop in cell power output due to exposure to 50 ppm  $\text{H}_2\text{S}$  was 32.3%, 18.5%, and 15.5% when cell voltage was 0.8, 0.6, and 0.3 V, respectively (the corresponding current density before sulfur poisoning was 115, 252, and 513  $\text{mA cm}^{-2}$ ) [18].

Thus, if the degree of sulfur poisoning is defined as the drop in cell power output (i.e., the drop in cell current under the potentiostatic mode or the drop in cell voltage under the galvanostatic mode), there is an apparent contradiction. On one hand, the relative drop in performance due to sulfur poisoning appears to be greater with increasing current density (or decreasing cell-terminal voltage) under galvanostatic conditions [2,14,15,20]; on the other hand, the relative drop in performance due to sulfur poisoning appears to be smaller with decreasing cell voltage (or

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increasing cell current density) under potentiostatic conditions [18,20]. Such an apparent contradiction has led to some confusion. For example, it is not clear whether an SOFC should be operated at higher current density (at lower voltage) or lower current density (at higher voltage) to alleviate the sulfur poisoning effect. In this paper, we provide an explanation for this apparent contradiction, which is further validated by experimental results. The significance of the analysis to the interpretation of sulfur poisoning/regeneration mechanism for Ni-based anodes of SOFCs is also discussed.

## 2. Experimental

The detailed procedures for cell fabrication and testing are as described elsewhere [18]. To measure the relative drop in cell power output, the cell was operated in either galvanostatic or potentiostatic mode, and the cell voltage (in the galvanostatic mode) or the cell current (in the potentiostatic mode) was monitored continuously as a hydrogen fuel containing a predetermined concentration of  $\text{H}_2\text{S}$  was introduced. Normally, the initial sulfur poisoning process of an electrolyte-supported cell would complete in approximately 5 min [13,16,18]. After that,  $\text{H}_2\text{S}$  was removed from the fuel stream and the cell power output gradually recovered to the original level. (The poisoning process was fully reversible since the time exposed to  $\text{H}_2\text{S}$  was very short [13,16,18].) The relative drop in cell power output is calculated as the ratio of the drop in cell voltage (or current) to the initial voltage (or current) value. To compare the result of cell impedance measurement with that of cell power output measurement, the impedance spectrum of the cell in clean hydrogen fuel under a dc bias (constant current or voltage) was measured. Then the cell was operated under the same dc bias while  $\text{H}_2\text{S}$  was introduced into the fuel stream. After the quick poisoning process completed, the dc measurement was stopped and the impedance spectrum was measured again under the same constant current or constant voltage conditions.

## 3. Results

### 3.1. Relative cell power output drop under constant current and constant voltage conditions

Fig. 1(a) shows the dependence of measured relative drops in cell power output on the concentration of  $\text{H}_2\text{S}$  when a cell was tested under a constant current density of 241 and 409  $\text{mA cm}^{-2}$ , respectively. The measured relative power output drop values are smaller than those reported in literature for similar electrolyte-supported cells: Xia and Birss reported that 10 ppm  $\text{H}_2\text{S}$  caused a cell voltage drop of 19.5% and 56.4% when the current density was 130 and 400  $\text{mA cm}^{-2}$ , respectively, at 800 °C [14]; Sasaki et al. reported cell failure (i.e., 100% voltage drop) for a cell operated at a constant current density of 200  $\text{mA cm}^{-2}$  at 850 °C when it was exposed to 5 ppm  $\text{H}_2\text{S}$  [13,16]. Nevertheless, the observed trend is generally consistent with what has been reported in the literature [2,14,15,20], that is, the measured relative drop in cell power output caused by sulfur poisoning was

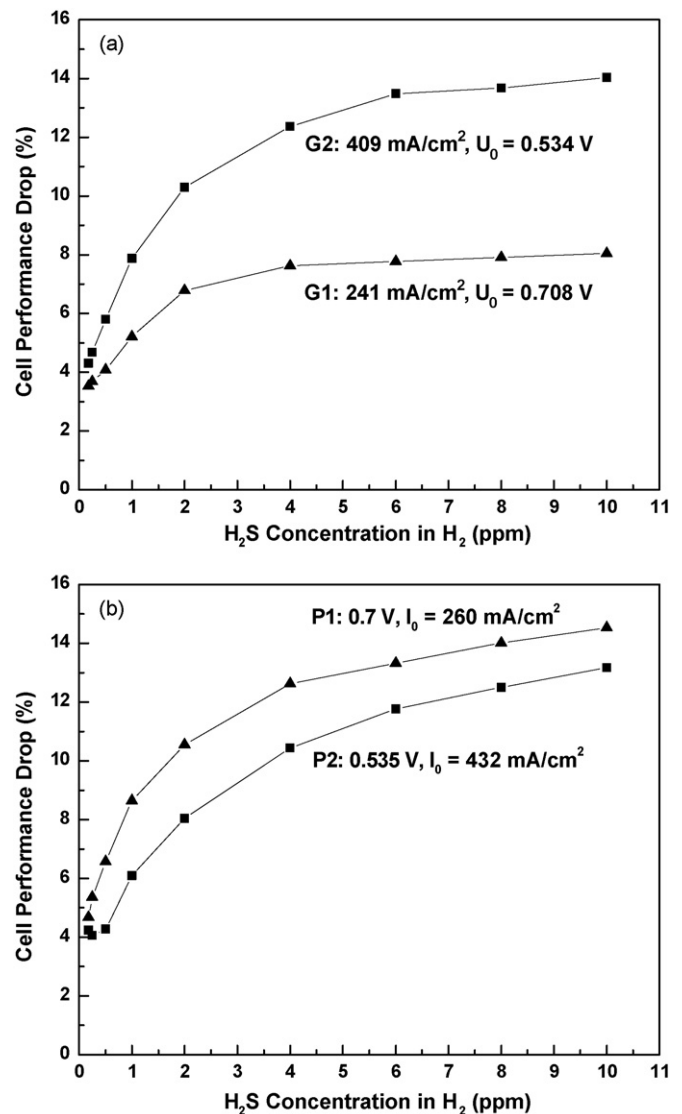


Fig. 1. Measured relative cell power output drop vs.  $p\text{H}_2\text{S}/p\text{H}_2$  for cells operated under galvanostatic (a) and potentiostatic (b) conditions at 800 °C.

always greater when more current was drawn from the cell for all concentrations of  $\text{H}_2\text{S}$  tested.

In contrast, Fig. 1(b) shows plots of measured relative drops in power output as a function of  $p\text{H}_2\text{S}/p\text{H}_2$  when the cell was tested under a constant cell voltage of 0.535 and 0.7 V, respectively. Contrary to the results obtained under constant current conditions but consistent with our previous study [18] and the study by Sprenkle et al. [20], the measured relative drop in cell power output caused by sulfur poisoning was smaller as the cell-terminal voltage was reduced from 0.7 to 0.535 V (the corresponding cell current density before sulfur poisoning was 432  $\text{mA cm}^{-2}$  for 0.535 V and 260  $\text{mA cm}^{-2}$  for 0.7 V).

### 3.2. Equivalent circuit analysis

This apparent contradiction can be explained using a simple equivalent circuit analysis. For the case of sulfur poisoning under a constant current (galvanostatic) condition, the cell current,  $I$ ,

is kept constant, i.e.,  $I=I_0$ . Before switching from  $H_2$  to  $H_2S$ -contaminated  $H_2$ , the cell-terminal voltage,  $U_0$ , is given by

$$U_0 = E_0 - I_0 R_{\text{cell},0}, \quad (1)$$

where  $E_0$  is the open circuit voltage and  $R_{\text{cell},0}$  is the total cell resistance before sulfur poisoning. After the cell is poisoned by  $H_2S$ , the cell total internal resistance changes from  $R_{\text{cell},0}$  to  $R_{\text{cell},0} + \Delta R_{\text{cell}}$ , and the cell-terminal voltage changes to

$$U = E_0 - I_0(R_{\text{cell},0} + \Delta R_{\text{cell}}). \quad (2)$$

The relative decrease in cell power output,  $\Delta P_r$ , is

$$\begin{aligned} \Delta P_r &= \frac{I_0(U_0 - U)}{I_0 U_0} \\ &= \frac{(E_0 - I_0 R_{\text{cell},0}) - [E_0 - I_0(R_{\text{cell},0} + \Delta R_{\text{cell}})]}{U_0} \\ &= \frac{I_0 \Delta R_{\text{cell}}}{U_0}. \end{aligned} \quad (3)$$

Considering that

$$I_0 = \frac{E_0 - U_0}{R_{\text{cell},0}}, \quad (4)$$

We can rewrite Eq. (3) as

$$\Delta P_r = \frac{E_0 - U_0}{R_{\text{cell},0}} \frac{\Delta R_{\text{cell}}}{U_0} = \frac{\Delta R_{\text{cell}}}{R_{\text{cell},0}} \frac{E_0 - U_0}{U_0}. \quad (5)$$

Eq. (5) shows that, in the galvanostatic (constant current) mode, the measured relative drop in cell power output (or in voltage),  $\Delta P_r$ , is a function of three variables: the relative increase in cell resistance ( $\Delta R_{\text{cell}}/R_{\text{cell},0}$ ), the initial cell-terminal voltage ( $U_0$ ), and the open cell voltage ( $E_0$ ). From Eq. (5), it is clear that  $\Delta P_r < \Delta R_{\text{cell}}/R_{\text{cell},0}$  when  $U_0 > 0.5E_0$  while  $\Delta P_r > \Delta R_{\text{cell}}/R_{\text{cell},0}$  when  $U_0 < 0.5E_0$ . Rearrange Eq. (5) and define  $\Delta R_{\text{cell},r} = \Delta R_{\text{cell}}/R_{\text{cell},0}$ , we have

$$\Delta R_{\text{cell},r} = \frac{\Delta R_{\text{cell}}}{R_{\text{cell},0}} = \Delta P_r \frac{U_0}{E_0 - U_0}. \quad (6)$$

Although we are unable to mathematically predict the trend of relative increase in cell resistance ( $\Delta R_{\text{cell},r}$ ) directly from the observed trend in relative cell performance drop ( $\Delta P_r$ ), we can calculate the  $\Delta R_{\text{cell},r}$  values using the  $E_0$ ,  $U_0$ , and  $\Delta P_r$  values measured in the experiment and examine the trend.

On the other hand, when the cell is tested in a potentiostatic mode, the cell-terminal voltage  $U$  is kept constant, i.e.,  $U=U_0$ . Before sulfur poisoning, the current passing through the cell is given by

$$I_0 = \frac{E_0 - U_0}{R_{\text{cell},0}}. \quad (7)$$

Upon poisoning by  $H_2S$ , the cell internal resistance changes from  $R_{\text{cell},0}$  to  $R_{\text{cell},0} + \Delta R_{\text{cell}}$ , and the cell current changes to

$$I = \frac{E_0 - U_0}{R_{\text{cell},0} + \Delta R_{\text{cell}}}. \quad (8)$$

The relative decrease in cell power output,  $\Delta P_r$ , is

$$\begin{aligned} \Delta P_r &= \frac{U_0(I_0 - I)}{U_0 I_0} \\ &= \frac{((E_0 - U_0)/R_{\text{cell},0}) - (E_0 - U_0)/(R_{\text{cell},0} + \Delta R_{\text{cell}})}{(E_0 - U_0)/R_{\text{cell},0}} \\ &= \frac{\Delta R_{\text{cell}}}{R_{\text{cell},0} + \Delta R_{\text{cell}}}, \end{aligned} \quad (9)$$

or

$$\Delta P_r = \frac{\Delta R_{\text{cell}}/R_{\text{cell},0}}{1 + \Delta R_{\text{cell}}/R_{\text{cell},0}}. \quad (10)$$

Eq. (10) shows that the relative drop in cell power output (or drop in current in this case),  $\Delta P_r$ , depends only on the relative increase in cell internal resistance ( $\Delta R_{\text{cell},r} = \Delta R_{\text{cell}}/R_{\text{cell},0}$ ) under the potentiostatic condition.  $\Delta P_r$  will always be smaller than  $\Delta R_{\text{cell},r}$ . Based on Eq. (10), the relative change in cell internal resistance,  $\Delta R_{\text{cell},r}$ , could also be calculated from the relative change in cell performance:

$$\Delta R_{\text{cell},r} = \frac{\Delta R_{\text{cell}}}{R_{\text{cell},0}} = \frac{\Delta P_r}{1 - \Delta P_r} = \frac{1}{1 - \Delta P_r} - 1. \quad (11)$$

Eq. (11) allows for the calculation of the relative increase in cell resistance due to sulfur poisoning ( $\Delta R_{\text{cell},r}$ ) from the observed relative performance loss; it also indicates the relative increase in cell resistance ( $\Delta R_{\text{cell},r}$ ) is a monotonous function of the relative drop in cell performance, implying that the smaller the  $\Delta P_r$ , the smaller the  $\Delta R_{\text{cell},r}$ . The later is important in that we can infer that the relative increase in cell resistance is also smaller when the cell current is higher under the potentiostatic condition.

Using the relationship obtained above for sulfur poisoning under galvanostatic and potentiostatic modes, we can now evaluate whether the sulfur poisoning behavior is consistent in terms of the relative increase in cell resistance under both conditions. Fig. 2 shows plots of the relative cell resistance increase calculated from the data in Fig. 1 using Eqs. (6) and (11) as a function of  $H_2S$  concentration for both galvanostatic and potentiostatic conditions. Clearly, the relative increase in cell resistance ( $\Delta R_{\text{cell},r}$ ) is smaller when the cell current is higher or the cell-terminal voltage is lower (closer to short circuit condition) under both conditions.

### 3.3. Verification using impedance measurements

To further verify the result from the equivalent circuit analysis described above, experiments were carried out to directly measure the change in cell resistance using impedance spectroscopy before and after sulfur poisoning under both galvanostatic and potentiostatic conditions. Shown in Figs. 3 and 4 are some typical impedance spectra. Table 1 summarizes the relative losses in power output and the corresponding relative increases in cell resistance, both experimentally measured and calculated using Eqs. (6) or (11). Under both galvanostatic and potentiostatic conditions, the relative increase in cell resistance ( $\Delta R_{\text{cell},r}$ ) is indeed smaller when the cell current density is higher (or when

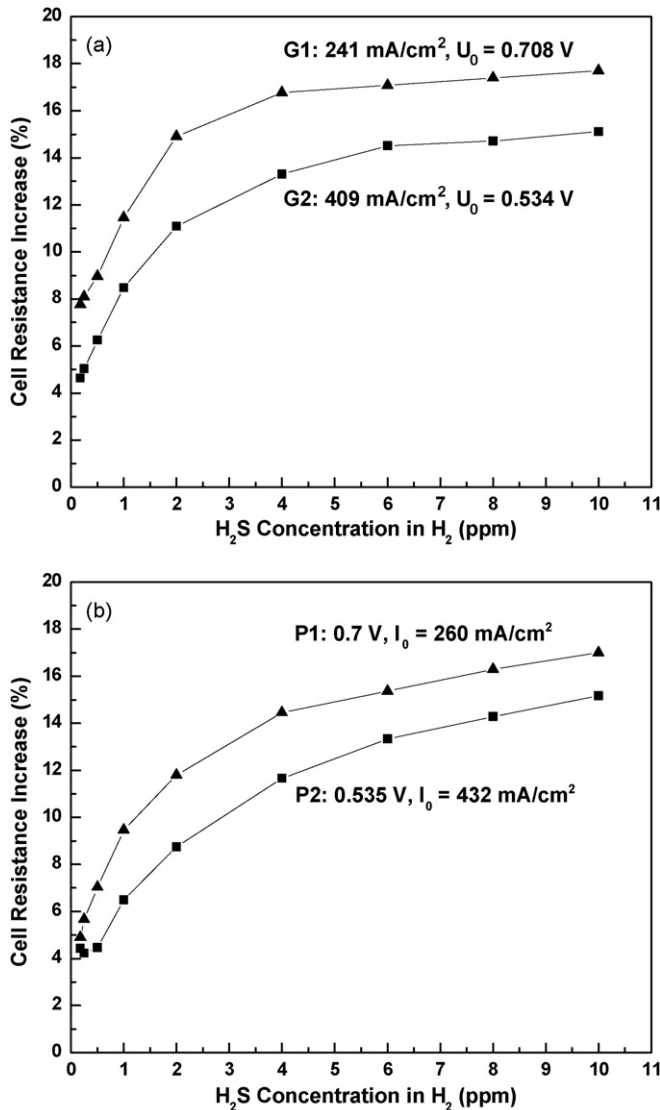


Fig. 2. Calculated relative cell power output drop vs.  $p\text{H}_2\text{S}/p\text{H}_2$  for cells operated under galvanostatic (a) and potentiostatic (b) conditions using the data in Fig. 1 at  $800^\circ\text{C}$ .

the cell-terminal voltage is lower), even though the measured relative drops in cell power show an opposite trend when measured under constant voltage conditions versus constant current conditions.

#### 4. Discussion

The results shown in Figs. 2–4 and Table 1 indicate that the relative increase in total cell internal resistance is smaller when the cell current density is greater or when the cell-terminal voltage is lower. The apparent contradiction seen for the trend in cell power output drop with respect to changes in cell current density or voltage can be explained as follows. Under both testing conditions, the anode polarization resistance increases with the degree of sulfur poisoning. To maintain a constant cell current or terminal voltage, however, the effective resistance for the external circuit (i.e., the potentiostat/galvanostat used) has to be adjusted accordingly during the test. For example, in the galvanostatic mode, the effective external circuit resistance has to be reduced in order to maintain a constant cell current. Similarly, in the potentiostatic mode, the effective external circuit resistance has to be increased to maintain a constant cell-terminal voltage. Both these effects are modulations of the original sulfur poisoning behavior, as represented by the relative increase in cell resistance, which leads to the difference in the observed drop in power output.

Nevertheless, the measurements under constant cell current or voltage provide useful information on sulfur poisoning. Measurements under potentiostatic conditions have the advantage that the observed trend in current change is consistent with that in the actual cell resistance change. However, the cell current drops as the anode is poisoned by sulfur, leading to an increase in cathode polarization resistance but a decrease in cathode overpotential, which increases the difficulty in analyzing the kinetic information. By comparison, in the galvanostatic mode, because we can assume that the cathode process does not change much (at least during the quick poisoning process), the measured increase in cell-terminal potential and cell resistance could be directly regarded as the increase in anode overpotential and polarization resistance. Thus, unless the cathodic polarization can be separated from the anodic polarization, galvanostatic (constant

Table 1

Relative performance loss,  $\Delta P_r$ ; relative cell resistance increase,  $\Delta R_{\text{cell},r}$  (both calculated using Eqs. (6) and (11) and experimentally measured); and relative cell interfacial resistance increase,  $\Delta R_{i,r}$ ; for the poisoning of Ni-YSZ anode caused by 10 ppm  $\text{H}_2\text{S}$  under constant current and constant voltage conditions

$i$ ( $\text{mA cm}^{-2}$ )	$U_0$ (V)	$\Delta P_r$ (%)	Calculated $\Delta R_{\text{cell},r}$ (%)	Measured $\Delta R_{\text{cell},r}$ (%)	Measured $\Delta R_{i,r}$ (%)
44	0.947	4.3	29.1	21.7	36.2
129	0.756	7.7	17.6	15.2	26.9
258	0.569	13.6	15.0	7.9	18.2
391	0.376	21.8	11.6	3.0	8.0
$U$ (V)	$i_0$ ( $\text{mA cm}^{-2}$ )	$\Delta P_r$ (%)	Calculated $\Delta R_{\text{cell},r}$ (%)	Measured $\Delta R_{\text{cell},r}$ (%)	Measured $\Delta R_{i,r}$ (%)
0.90	44	31.5	46.0	29.2	50.0
0.70	144	22.0	28.2	20.0	50.0
0.50	246	15.7	18.7	13.3	38.7
0.35	314	12.6	14.4	4.5	18.5

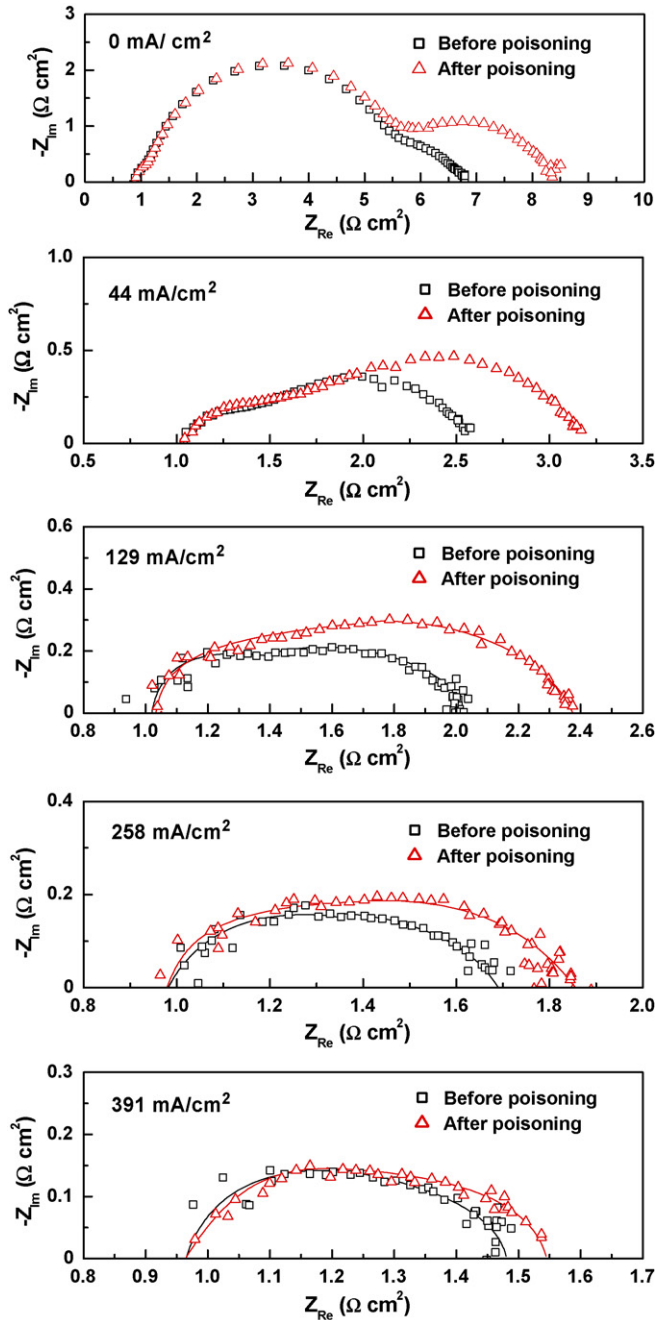


Fig. 3. Comparison of impedance spectra for an electrolyte-supported cell operated at a constant current density of 0, 44, 129, 258, and 391 mA cm<sup>-2</sup> before and after 10 ppm H<sub>2</sub>S was introduced into the fuel flow at 800 °C.

current) mode is better for impedance and cell power output measurements.

While it is still not clear why a smaller increase in cell internal resistance was observed at a higher cell current density (or lower cell voltage), it is most likely related to the removal of the adsorbed sulfur atoms. This is because most previous studies proposed that the poisoning of SOFC anodes by ppm level H<sub>2</sub>S is due to the blockage of surface active sites by adsorbed sulfur atoms [9,11,15,16,18,19]. Previously, it has been suggested that the adsorbed sulfur could be electrochemically oxidized by oxygen ions at triple phase boundary (TPB) into SO<sub>2</sub> as

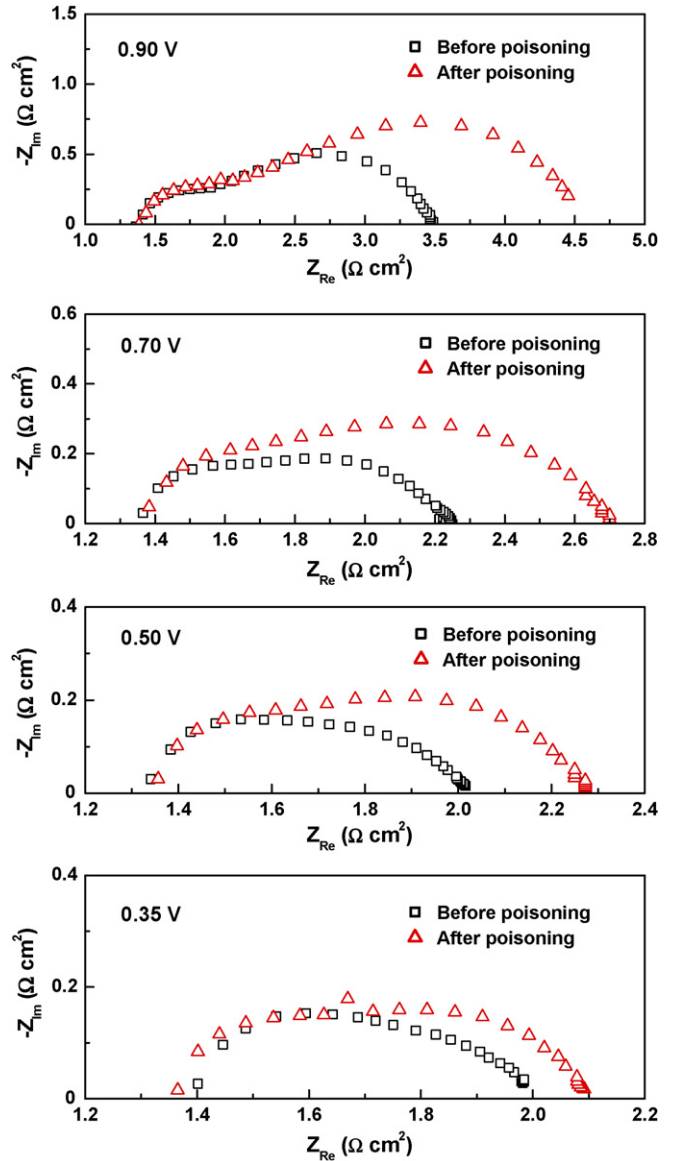
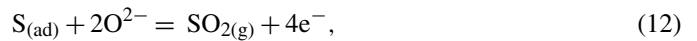


Fig. 4. Comparison of impedance spectra for an electrolyte-supported cell operated at a constant voltage of 0.90, 0.70, 0.50, and 0.35 V before and after 10 ppm H<sub>2</sub>S was introduced into the fuel flow at 800 °C.

follows [18]



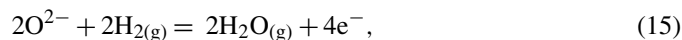
SO<sub>2</sub> desorbs from the surface, leading to alleviated poisoning effect. Such an explanation seems reasonable since one would expect that higher oxygen ion flux would lead to quicker removal of more adsorbed sulfur. It is noted that SO<sub>2</sub> may exist only as a “short-lived” reaction intermediate because SO<sub>2</sub> may be reduced back to H<sub>2</sub>S in the fuel environment (at least from a thermodynamic point of view). For example, for a humidified hydrogen with  $p_{H_2O}/p_{H_2} = 3:97$ , the equilibrium  $p_{SO_2}/p_{H_2S}$  ratio would be on the order of  $\sim 6 \times 10^{-11}$  at 1000 K for the reaction of



In fact, if we combine reactions (12) and (13) and rearrange the equations, we have



and



which are equivalent to the desorption of sulfur in an  $H_2S/H_2$  environment on the electrode (reaction (14)) plus the electrochemical oxidation of  $H_2$  (reaction (15)).

## 5. Conclusions

The degree of sulfur poisoning of Ni-YSZ cermet anode is most accurately described by the increase in cell anode polarization resistance due to sulfur poisoning. It could be misleading to use the relative drop in performance (or in power output) to characterize the degree of sulfur poisoning. The contradictory trends in performance loss due to sulfur poisoning observed under galvanostatic and potentiostatic conditions were explained using an equivalent circuit analysis. The degree of sulfur poisoning, if measured by the increase in cell internal resistance, is smaller when more current is drawn from the cell in either galvanostatic or potentiostatic conditions. The observed trend seems consistent with the proposed mechanism for sulfur poisoning/regeneration process in which more oxygen ion passing through the electrolyte alleviates the poisoning process.

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