



## High-Performance Anode for H<sub>2</sub>S-Air SOFCs

Guo-Lin Wei, Jing-Li Luo,\*<sup>z</sup> Alan R. Sanger, and Karl T. Chuang

Department of Chemical and Materials Engineering, University of Alberta, Edmonton, Alberta T6G 2G6, Canada

A high-performance composite anode has been developed for H<sub>2</sub>S-air solid oxide fuel cells (SOFCs). The anode design is based on the requirements of three-phase boundary theory. The anode material comprises a mixture of composite metal sulfide (Mo-Ni-S) catalyst prepared from MoS<sub>2</sub> and NiS (1:1 weight ratio), admixed with up to 10% each of Ag as electronic conductor and yttria-stabilized zirconia (YSZ) as ionic conductor. The optimum composition is about 90 wt % Mo-Ni-S, 5 wt % Ag, and 5 wt % YSZ. A fuel cell using a 0.2 mm thick YSZ membrane produced a maximum sustainable current density over 480 mA cm<sup>-2</sup> at 750°C and over 800 mA cm<sup>-2</sup> at 850°C, and maximum power density 50 mW cm<sup>-2</sup> at 750°C and over 200 mW cm<sup>-2</sup> at 850°C. © 2004 The Electrochemical Society. [DOI: 10.1149/1.1636177] All rights reserved.

Manuscript submitted March 27, 2003; revised manuscript received July 10, 2003. Available electronically January 8, 2004.

H<sub>2</sub>S is a severe pollutant that is a by-product of many industrial operations such as processing of natural gas, coking, and hydrodesulfurization of crude oil or coal.<sup>1</sup> It is anticipated that additional large quantities of H<sub>2</sub>S will be produced when coal liquefaction attains commercial importance.<sup>1</sup> Thus, H<sub>2</sub>S is not only a major pollutant, but is also an increasingly significant resource for sulfur production.

Many processes have been developed to remove and recover H<sub>2</sub>S, including adsorption and absorption, conversion to elemental sulfur, and oxidation to sulfur oxides. The capital investment required for construction of sulfur recovery facilities is significant. There is increasing pressure to maximize recovery of H<sub>2</sub>S to minimize its environmental impact, and throughput at minimum investment is critical to the chemical process industry. Fuel-cell technology provides a potentially economical and powerful alternative for sulfur recovery. It can generate high-grade electric power from the large amount of chemical energy available from the oxidation of H<sub>2</sub>S<sup>2</sup> and can convert H<sub>2</sub>S selectively to high-purity sulfur. Since the first reports of direct H<sub>2</sub>S-air solid oxide fuel cells (SOFCs) by Sammells *et al.*,<sup>2-4</sup> several attempts have been made to improve cell performance.<sup>5-11</sup> Currently, yttria-stabilized zirconia (YSZ) is the most commonly used oxide ion-conducting electrolyte, and either Pt<sup>5,6,8-11</sup> or strontium-doped lanthanum manganite (LSM)<sup>2-4,7</sup> is used as cathode catalyst. Because of the highly corrosive atmosphere in the anode chamber of H<sub>2</sub>S-air SOFCs, the choice of anode material is limited primarily to metal sulfides, such as thiospinels,<sup>2-4</sup> WS<sub>2</sub>,<sup>3,4,7</sup> CoS<sub>1.035</sub>, or Li<sub>2</sub>S,<sup>7</sup> or metal oxides that are converted into the corresponding sulfides in the high-temperature H<sub>2</sub>S atmosphere at the anode, such as LiCoO<sub>2</sub>, which is converted to Li<sub>2</sub>S/CoS<sub>1.035</sub>.<sup>7</sup> Thin (40 μm) YSZ electrolyte films are available, for example, as provided in a Siemens Westinghouse tubular SOFC system. Using these thin films, power densities over 412 mW cm<sup>-2</sup> (1302 mA cm<sup>-2</sup> at 0.317 V) have been obtained at 770°C with a Li<sub>2</sub>S/CoS<sub>1.035</sub>-based anode.<sup>7</sup> Power density of almost 90 mW cm<sup>-2</sup> and short-circuit current density near 300 mA cm<sup>-2</sup> were obtained at 830°C when using a thick (400-500 μm) YSZ disk of the same anode material.<sup>7</sup>

Three-phase boundary (TPB) theory lays some fundamental principles for the development of high-performance gas electrodes that are used in electrochemical devices, such as fuel cells and electrolyzers. According to TPB theory, ionic-conduction paths, electronic-conduction paths, and gas diffusion paths must all exist in a highly efficient gas electrode. Ideally, an electronic conductor, in most cases a metal or metal oxide, and a liquid or solid ionic conductor are both evenly distributed through the electrode. The electrode must be sufficiently porous to avoid any significant mass-transport

barrier. The value of TPB theory was demonstrated by its application in developing high-performance electrodes with significantly decreased Pt loading in proton exchange membrane fuel cells (PEMFCs), from 4 to 0.35 mg cm<sup>-2</sup>, with improved fuel cell performance.<sup>12</sup> Impregnation of Nafion electrolyte into electrode structures significantly extended the three-dimensional reaction zone and improved the catalyst efficiency for the same system. In H<sub>2</sub>-air SOFCs, it is also a common practice to admix YSZ into the composition of anode catalysts to increase the three-dimensional reaction zone. An LSM cathode is both oxide ion- and electron-conducting, and so there is no need to incorporate YSZ into this electrode material. However, to date the anode for H<sub>2</sub>S-air SOFCs usually consists of catalytic metal sulfides only, which are usually semiconductors at room temperature and to which no additional ionic conductor or electronic conductor has been added. This has had the effect of severely limiting the effective reaction zone and has resulted in very low catalyst efficiency.

In this work, we have applied TPB theory successfully to develop a high-performance metal sulfide-based anode for H<sub>2</sub>S-air SOFCs. This composite anode comprises a composite Mo-Ni-S metal sulfide catalyst derived from MoS<sub>2</sub> and NiS,<sup>10,11</sup> admixed with YSZ as ionic conductor and Ag as electronic conductor.

### Experimental

Anode materials of various compositions were prepared. The Mo-Ni-S composite sulfide was prepared by first mechanically mixing MoS<sub>2</sub> (Sigma-Aldrich, <2 μm) and NiS (Sigma-Aldrich, 150 mesh) in 1:1 weight ratio in a stirred solvent overnight. Then the mixture was dried at 110°C in an oven under ambient atmosphere. The resulting powder was then used to prepare further formulations by mixing the Mo-Ni-S composite sulfide with one or both of 0-10 wt % Ag powder (Sigma-Aldrich, 2-3.5 μm) and YSZ powder (Toso TZ-8Y) in the same solvent overnight. The solvent was evaporated. The resulting mixture was first dried in air at 110°C for 12 h and then heat-treated in nitrogen atmosphere at 1050°C for 2 h, followed by cooling down in the same nitrogen atmosphere to room temperature.

Details of the preparation of the PEN (positive-electrolyte-negative) assembly are in Ref. 10. Briefly, a 0.2 mm thick 8YSZ membrane (zirconia stabilized with 8% yttria, Intertec Southwest ZO-8Y) was used as electrolyte. Both cathode (Pt, Heraeus CL11-5100) and anode (paste in α-terpineol) were applied using the screen-printing technique. The cathode was applied first, and the cathode-membrane assembly was heated in air at 130°C for 30 min and then at 1050°C for a further 30 min. Then the anode was applied to the opposite face of the membrane, and the resulting assembly was heated in nitrogen atmosphere using the same procedure. The apparent electrode area was approximately 1 cm<sup>2</sup>. The thickness of the anode layer was varied between 50 and 70 μm as measured according to cross-sectional scanning electron microscopy (SEM) photos.

\* Electrochemical Society Active Member.

<sup>z</sup> E-mail: jingli.luo@ualberta.ca

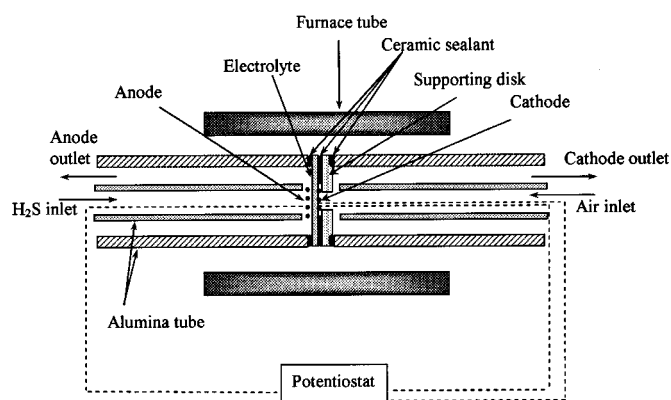


Figure 1. Test station for H<sub>2</sub>S-air SOFCs.

To install the PEN assembly into the fuel cell test station (Fig. 1), the cathode side first was attached to a supporting annular ceramic disk with ceramic sealant (Aremco 503) and dried in air for 30 min at room temperature. The supporting ceramic disk and attached PEN assembly were then sealed between two Al<sub>2</sub>O<sub>3</sub> peripheral tubes with the unattached side of the supporting disk facing the cathode chamber (Fig. 1). The sealant layer was 0.1–0.2 mm thick and 3 mm wide between the peripheral tubes and supporting disk/PEN assembly. An additional 3 mm wide sealant layer was applied outside the junctions with the Al<sub>2</sub>O<sub>3</sub> tubes and around the sealing area. Platinum mesh was used as current collector and was refreshed in the flame of a gas lamp before each experiment. The sealed cell was heated in a tubular furnace (Thermolyne F79300), with air passing through the cathode chamber and nitrogen through the anode chamber. To cure the sealant, the furnace temperature was increased slowly to 250°C, at which temperature it was kept for 1 h before being increased to 750°C for further curing.

To test cell performance, the anode feed was switched from nitrogen to pure H<sub>2</sub>S. After a stable open circuit voltage (OCV) had been obtained, cell current-voltage performance was measured with a Gamry electrochemical system (PC4-750), potentiodynamically and in the IR-compensation mode. Both the anode (H<sub>2</sub>S) and cathode (air) feeds were controlled at a flow rate of 25 mL/min with Matheson flow controllers. Electrochemical impedance spectrum (EIS) was obtained with Gamry system at stabilized OCV. The applied amplitude was 10 mV and frequency from 10<sup>5</sup> Hz to 0.2 Hz.

SEM and energy-dispersive X-ray spectrometry (EDX) measurements were conducted with a Hitachi S-2700 scanning electron microscope and PGT Imix system. X-ray diffraction (XRD) spectra were obtained using a Rigaku Rotaflex X-ray diffractometer.

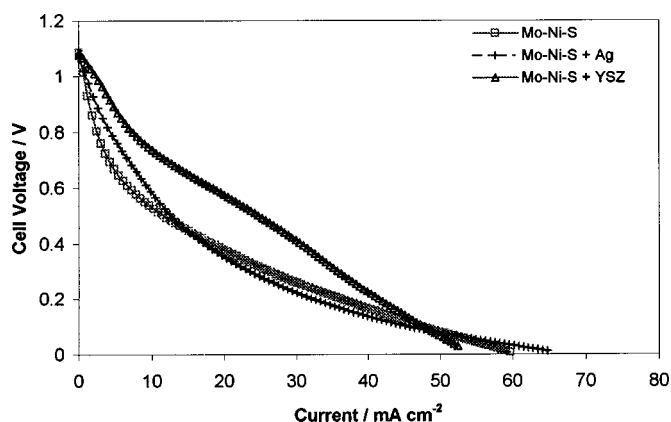


Figure 2. Current-voltage curves for H<sub>2</sub>S-air SOFCs with different anodes at 750°C.

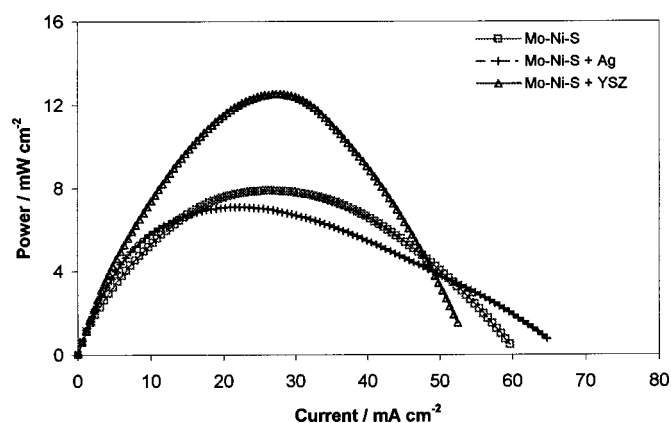


Figure 3. Current-power curves for H<sub>2</sub>S-air SOFCs with different anodes at 750°C.

## Results and Discussion

**Mo-Ni-S as anode catalyst.**—MoS<sub>2</sub> is a highly active catalyst in various hydrogenation/dehydrogenation and hydrodesulfurization processes used in the petroleum industry. It was also found to form an active anode catalyst that possessed good electronic conductivity, and a catalytic activity that was better than Pt in H<sub>2</sub>S-air SOFCs.<sup>13</sup> However, MoS<sub>2</sub> is volatile at high temperatures. In contrast, a Mo-Ni-S composite catalyst prepared from a mixture of MoS<sub>2</sub> and NiS has been found to be involatile.<sup>10</sup> Further, the presence of Ni enhanced the catalytic activity for hydrodesulfurization and related reactions.<sup>14</sup>

Baseline performance of the Mo-Ni-S catalyst was determined with an H<sub>2</sub>S-air SOFC using an anode comprising Mo-Ni-S alone. The current-voltage and current-power curves for operating temperatures of 750°C and for temperature of 850°C are shown in Fig. 2 and 3 and Fig. 4 and 5, respectively. In sequence, the current-voltage behavior was controlled by electrochemical reaction kinetics in the high-cell-voltage region (low electrode overpotentials); by reaction kinetics/mass transport in combination in the mid-cell-voltage region; and by mass transport at the low-cell-voltage region (high electrode overpotentials).<sup>10</sup> The data shown in Fig. 2 and 3 have been compensated for ohmic resistance polarization. At 750°C, the maximum current density was about 60 mA cm<sup>-2</sup>, and the maximum power density was 8 mW cm<sup>-2</sup> at a current density of 27 mA cm<sup>-2</sup> and cell voltage of 0.30 V. At 850°C, increased electrolyte conductivity and enhanced reaction rate improved the maximum current density to 99 mA cm<sup>-2</sup>. The maximum power density in-

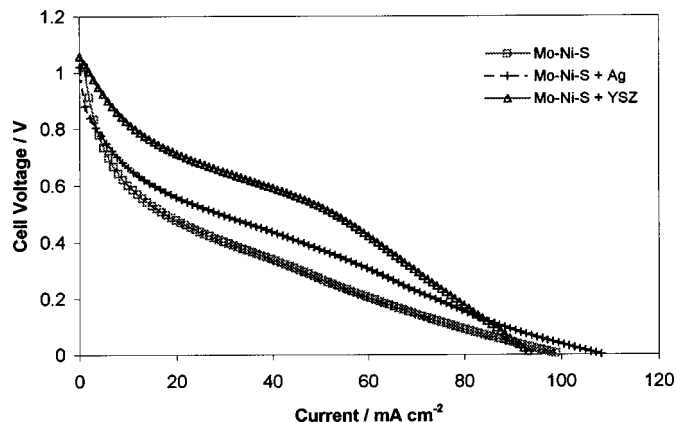
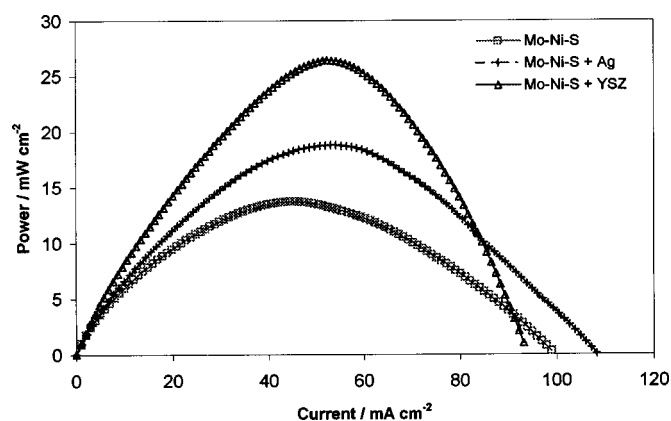


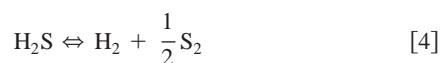
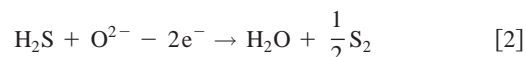
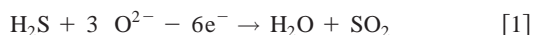
Figure 4. Current-voltage curves for H<sub>2</sub>S-air SOFCs with different anodes at 850°C.



**Figure 5.** Current-power curves for H<sub>2</sub>S-air SOFCs with different anodes at 850°C.

creased to about 14 mW cm<sup>-2</sup> at current density of 45 mA cm<sup>-2</sup> and cell voltage of 0.31 V. Cell voltage corresponding to the highest power density increased slightly when the operating temperature was increased from 750 to 850°C, as reported earlier.<sup>10</sup>

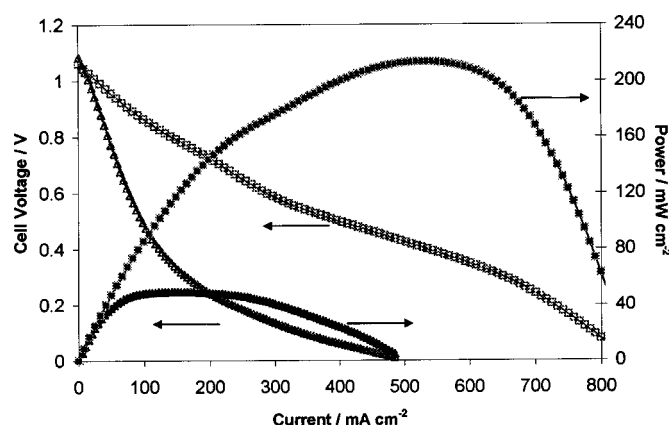
There are several reactions that may occur at the anode.<sup>6,10</sup> H<sub>2</sub>S can be converted directly to SO<sub>2</sub> (Eq. 1) or to elemental sulfur (Eq. 2). Alternatively, H<sub>2</sub> may be formed by internal reforming of H<sub>2</sub>S at temperatures in excess of 700°C according to Eq. 4. However, the equilibrium amount of H<sub>2</sub> formed by thermal decomposition of H<sub>2</sub>S (Eq. 4) is low in the present temperature range (less than 10%).



When the anode was Mo-Ni-S alone, it contained no strong ionic conductors. Consequently, anode reactions were limited to a zone closely adjacent to the electrolyte. MoS<sub>2</sub> and NiS are both semiconductors and the electronic resistivity of Mo-Ni-S catalyst is measured to be 0.51 Ω cm at 750°C and 1.01 Ω cm at 850°C, respectively.<sup>15</sup> The cell performance with this anode catalyst showed that it was capable of sustaining low electronic current densities.

**Cell performance as a function of anode composition.**—Admixtures were prepared by mixing 0–10 wt % of one or both Ag and YSZ to Mo-Ni-S. Ag was added as an electronic conductor and YSZ as an ionic conductor. It was found that addition of less than about 5 wt % of either Ag or YSZ produced little benefit. However, inclusion of over 10 wt % of either Ag or YSZ showed detrimental effects. Ag particles agglomerated on the anode surface when mixtures containing over 10 wt % Ag were heat-treated. Dense anode materials were produced when mixtures containing at least 10 wt % YSZ were heat-treated. These dense materials had lower current density and power density, probably due to increased mass-transfer resistance. It was found that the optimum amount of either Ag or YSZ was about 5 wt %, and so anodes with the formulations [95 wt % (Mo-Ni-S) + 5 wt % Ag], [95 wt % (Mo-Ni-S) + 5 wt % YSZ], and [90 wt % (Mo-Ni-S) + 5 wt % Ag + 5 wt % YSZ] were prepared for evaluation of the roles of each component.

Addition of 5 wt % Ag to Mo-Ni-S anode provided very little improvement in cell performances at 750°C (Fig. 2 and 3). The maximum values for current density (65 mA cm<sup>-2</sup>) and power den-



**Figure 6.** Performance of H<sub>2</sub>S-air SOFC with [90 wt % (Mo-Ni-S) + 5 wt % Ag + 5 wt % YSZ] anode at (△, ▲) 750 and (□, ■) 850°C.

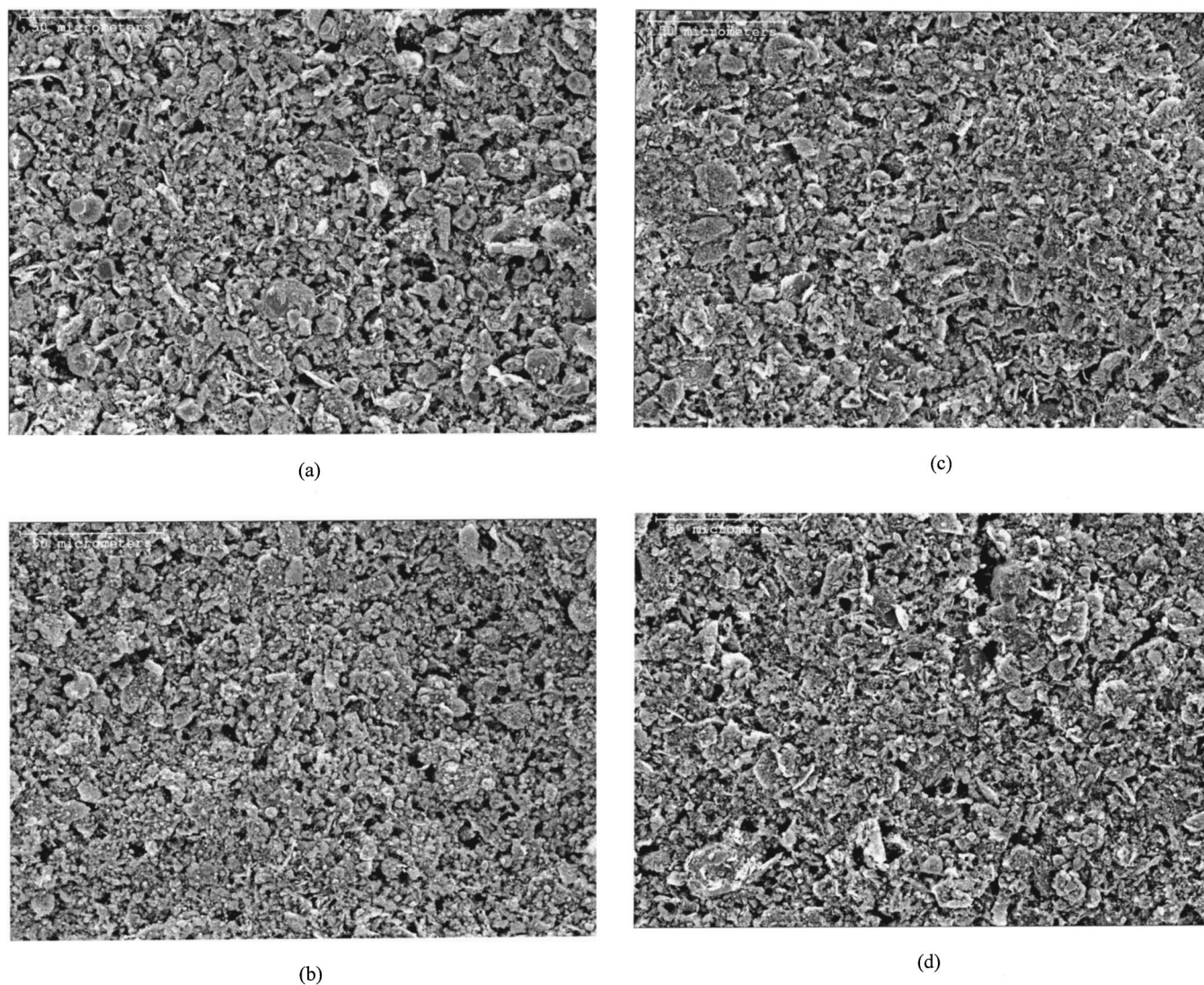
sity (7 mW cm<sup>-2</sup>) were within 10% of the values for Mo-Ni-S alone. This is understandable because Ag is an electronic conductor and did not enhance ionic conductivity in the anode, which was needed to increase the three-phase reaction zone in the anode and in turn to increase the overall cell performance. However, the maximum current density increased to 108 mA cm<sup>-2</sup> and the maximum power density to 19 mW cm<sup>-2</sup> at 850°C (Fig. 4 and 5). Similar improvements also were observed when Ag powder was admixed with Co-Mo-S anode catalyst.<sup>15</sup> As seen in Fig. 4 and 5, the improvement was more obvious in regions under mixing reaction kinetics/mass-transport control or pure mass-transport control, and so these data indicated an improvement in the mass-transport process.

Cell performance results with [Mo-Ni-S + YSZ] anode are shown in Fig. 2–5. At the same current density, fuel cells containing this anode showed higher voltages than anodes comprising Mo-Ni-S alone, in both the reaction-control region and the mixing reaction kinetics/mass-transport control region. In most of the mixing controlling region, the cell voltage was about 200 mV higher at 750°C and 150 mV higher at 850°C. The maximum power density was 13 mW cm<sup>-2</sup> at 28 mA cm<sup>-2</sup> and 0.45 V at 750°C, and 26 mW cm<sup>-2</sup> at 50 mA cm<sup>-2</sup> and 0.5 V at 850°C. According to TPB theory, mixing of Mo-Ni-S with oxide ion-conducting YSZ would be expected to increase the reaction zone in the anode and thereby improve the overall cell performance. In this case, YSZ provides extended oxide-conducting paths in the anode and thereby expands the reaction zone throughout the electrode. The cell performance results showed in Fig. 2–5 support this proposition.

However, anode morphology also played a role in cell performances. Cell voltages with the [Mo-Ni-S + YSZ] anode decreased more quickly in the mass-transport-control region than cells with a Mo-Ni-S anode. As seen below, anode particles were enlarged or agglomerated with the average particle size of 13.4 μm. The enlarged and agglomerated anode particles imposed an extra barrier for mass-transport processes of reactants and products, and therefore inhibited further increase in cell current and power.

A composite anode was designed comprising an admixture of Mo-Ni-S, Ag, and YSZ powders [95:5:5, weight ratio]. The overall cell performance showed significant improvement and cell voltage was much higher at both 750 and 850°C (Fig. 6). The maximum current density was over 480 mA cm<sup>-2</sup> at 750°C and over 800 mA cm<sup>-2</sup> at 800°C. The maximum power density was 50 mW cm<sup>-2</sup> at 150 mA cm<sup>-2</sup> and 0.34 V at 750°C and 213 mW cm<sup>-2</sup> at 536 mA cm<sup>-2</sup> and 0.4 V at 850°C (Fig. 6). These values were significantly higher than those reported for cells having Li<sub>2</sub>S-CoS<sub>1.035</sub> anodes and a thicker YSZ membrane.<sup>7</sup> It also is seen later that the anode morphology was improved in comparison with that of either Mo-Ni-S alone or Mo-Ni-S admixed with any one of Ag or YSZ. The average





**Figure 7.** SEM photos of anode catalysts before cell performance test: (a) Mo-Ni-S alone; (b) [(Mo-Ni-S) + Ag]; (c) [(Mo-Ni-S) + YSZ]; and (d) [(Mo-Ni-S) + Ag + YSZ].

particle size of this anode was 9.1  $\mu\text{m}$ . In accordance with TPB theory, mixing of Mo-Ni-S with both oxide ion-conducting YSZ and electron-conducting Ag further increased the reaction area to a greater distance from the electrolyte surface and thereby greatly improved the cell performance.

A barrier to performance was imposed by mass-transport-control processes, especially in the high-current-density region at 850°C (Fig. 6), as also found for the [Mo-Ni-S + YSZ] anode (Fig. 2-5). Thus, the microstructure of anode materials must be modified to achieve further improvement in current and power density.

**Anode morphology.**—The association between fuel cell performance and anode morphology made previously was supported by the corresponding morphological differences observed in SEM photos of the newly prepared anodes (Fig. 7a-d). All anodes were porous but slightly sintered. However, the average particle size in the [Mo-Ni-S + Ag] anode (9.5  $\mu\text{m}$ , Fig. 7b) was smaller than that in the Mo-Ni-S anode (12.5  $\mu\text{m}$ , Fig. 7a). Similarly, the average particle size for [Mo-Ni-S + Ag + YSZ] (9.1  $\mu\text{m}$ , Fig. 7d) was less than that for [Mo-Ni-S + YSZ] (13.4  $\mu\text{m}$ , Fig. 7c). The anodes containing Ag each were less sintered, had higher surface area, and were more porous than the corresponding anodes without Ag. Thus, Ag plays a significant role in determining the macrostructure of the

anode. The original Ag particle size was 2-3.5  $\mu\text{m}$ . The melting point of Ag is 961.8°C, and so Ag particles melted during heat-treatment during preparation of the anode material. EDX examination of the anode admixtures so prepared showed that Ag was distributed throughout the entire electrode structure. The SEM and EDX results for [Mo-Ni-S + Ag] suggested that Ag was distributed over the surfaces of larger Mo-Ni-S particles during heat-treatment and formed distributed fine particles after the mixture was cooled below the melting point of Ag. The smaller sizes of Mo-Ni-S particles found for the admixtures containing Ag compared to those without Ag suggest that small Ag particles that adhered onto Mo-Ni-S inhibited sintering of the latter to form larger particles. No significant morphology changes were observed (SEM) when comparing fresh and used anodes.

A problem previously identified for use of  $\text{Li}_2\text{S-CoS}_{1.035}$  anode materials was poor adherence of the anode to the electrolyte.<sup>7</sup> It has now been found that adherence is improved by incorporation of YSZ in the anode formulation. Cell performance was stable with time for a sequence of tests conducted over several days. In contrast, a slow degradation in performance was reported by Yates and Winnick for a fuel cell using  $\text{Li}_2\text{S-CoS}_{1.035}$  anode and a thicker YSZ membrane, and even worse degradation occurred when using a fuel

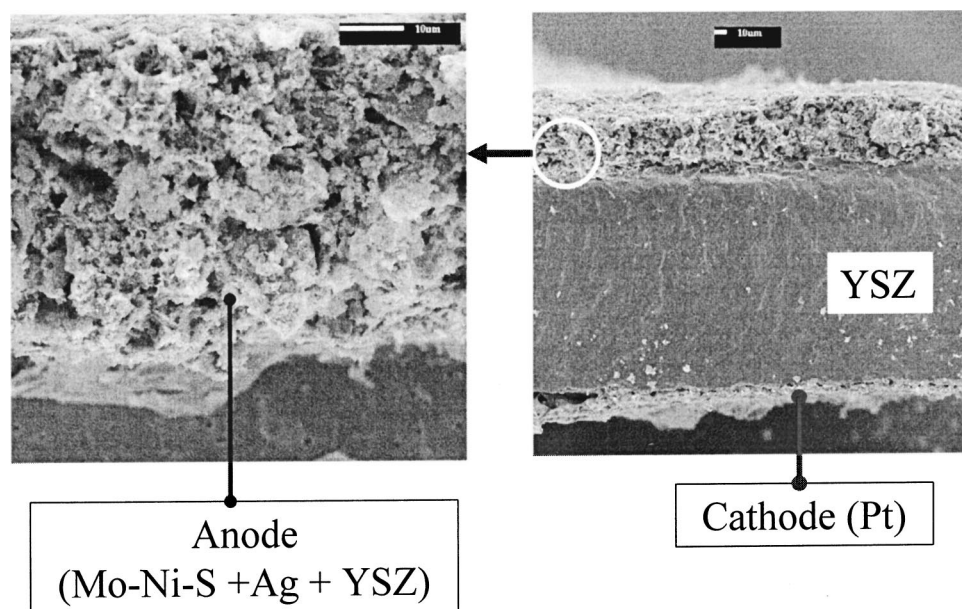


Figure 8. Cross-sectional SEM photos of a tested PEN unit.

cell having a thin, tubular YSZ layer.<sup>7</sup> Slow delamination of their anode was identified as one of the causes of degradation. The stability of our anode is attributed to an improvement in anode-electrolyte bonding arising from the presence of YSZ in the anode formulation. The secure bonding of the anode cross the electrolyte surface is shown in the cross-sectional SEM photos of a tested PEN unit with a thickness of anode layer of *ca.* 60 µm (Fig. 8). The photos illustrated also the porous and homogeneous structure of the [90 wt % (Mo-Ni-S) + 5 wt % Ag + 5 wt % YSZ] anode, which was unchanged after cell performance tests.

It is well understood that Ag readily reacts with sulfur at low temperature to form Ag<sub>2</sub>S. This reaction is reversed at temperatures above 200°C when Ag<sub>2</sub>S thermally decomposes to its elements.<sup>16</sup> Therefore, whereas formation of PtS increased resistance and caused detachment of the current collector, there were no similar adverse effects resulting from use of Ag powder.<sup>15</sup> The XRD technique was used in comparing the compositions of fresh and used anode materials (Fig. 9). No new components were identified in used materials.

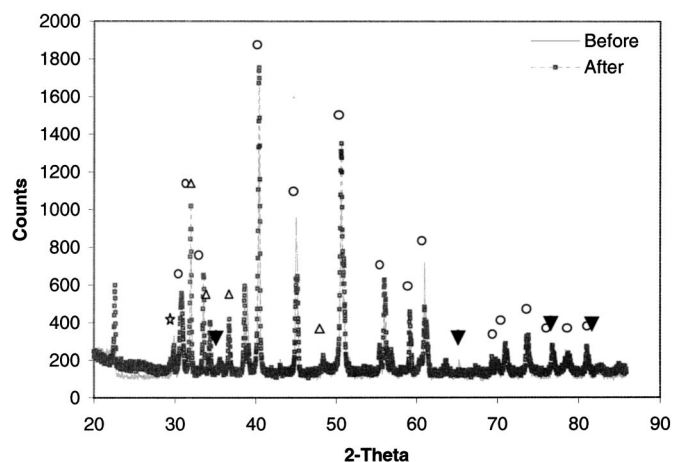


Figure 9. XRD spectra of fresh and tested [Mo-Ni-S + Ag + YSZ] anode material: (○) MoS<sub>2</sub>, (△) NiS, (▼) Ag, and (★) (Y<sub>2</sub>O<sub>3</sub>)<sub>0.15</sub>(ZrO<sub>2</sub>)<sub>0.85</sub>.

**Electrochemical impedance spectroscopy.**—EIS has been proved to be a useful tool in elucidating the electrode processes in fuel cells.<sup>17</sup> Figures 10 and 11 show the EIS for the different formulations of the anode materials at 750 and 850°C, respectively. As expected, increase in the operating temperature decreased both the ohmic and polarization resistances. At each temperature the ohmic resistance of the Mo-Ni-S and [Mo-Ni-S + YSZ] anodes were each significantly higher than the corresponding electrodes admixed with Ag. Thus, the improvement in electrical conductivity conferred by Ag is evident whereas there was no improvement when YSZ was admixed with Mo-Ni-S.

There is a parallel reduction in the charge-transfer resistance with the addition of Ag to Mo-Ni-S anode. Reduction in the charge-transfer resistance was also conferred with the addition of YSZ to Mo-Ni-S. In accordance with the current-voltage performances, the [90 wt % (Mo-Ni-S) + 5 wt % Ag + 5 wt % YSZ] anode showed significantly lower charge-transfer resistance. The admixture of both Ag and YSZ to Mo-Ni-S anodes provided complementary benefits to its electrochemical performance.

Several anodic reactions occurred simultaneously in the H<sub>2</sub>S-air fuel cell. Detailed investigations using the EIS technique may provide more useful information regarding the reaction mechanisms, which will be the subject of our future study.

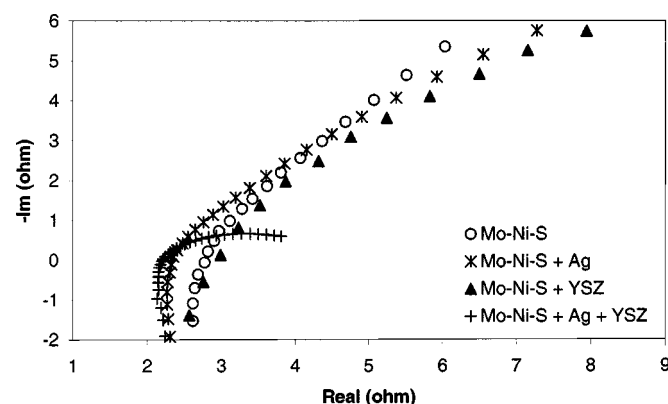


Figure 10. EIS of different anodes at 750°C.



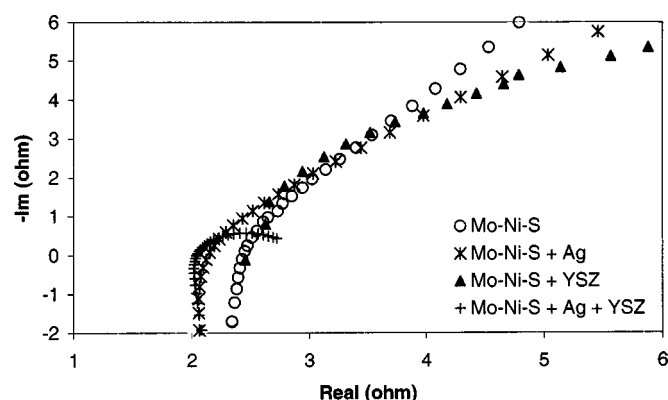


Figure 11. EIS of different anodes at 850°C.

### Conclusions

A high-performance anode has been developed based on TPB theory. The anode consists of an admixture of Mo-Ni-S with Ag and YSZ powders, with an optimum composition about 90:5:5 by weight. The maximum power density was  $50 \text{ mW cm}^{-2}$  at  $750^\circ\text{C}$  and over  $200 \text{ mW cm}^{-2}$  at  $850^\circ\text{C}$ . The anode materials were stable over time on stream.

It has been shown that inclusion of Ag in the formulation of the anode material reduced both the ohmic and reaction resistance. Inclusion of YSZ also reduced reaction resistance but had little effect on ohmic resistance.

### Acknowledgments

Financial support from a Natural Sciences and Engineering Research Council of Canada Strategic Project Grant is gratefully acknowledged.

The University of Alberta assisted in meeting the publication costs of this article.

### References

1. E. D. Weil and S. R. Sandler, in *Kirk-Othmer Encyclopedia of Chemical Technology* (on-line edition), John Wiley & Sons, New York (2002); <http://www.mrw.interscience.wiley.com/kirk/articles/sulfweil.a01/frame.html>
2. N. U. Pujare, K. W. Semkow, and A. F. Sammells, *J. Electrochem. Soc.*, **134**, 2639 (1987).
3. A. F. Sammells, U.S. Pat. 4,920,015 (1990).
4. N. U. Pujare, K. J. Tsai, and A. F. Sammells, *J. Electrochem. Soc.*, **136**, 3662 (1989).
5. T. J. Kirk and J. Winnick, *J. Electrochem. Soc.*, **140**, 3494 (1993).
6. D. Peterson and J. Winnick, *J. Electrochem. Soc.*, **145**, 1449 (1998).
7. C. Yates and J. Winnick, *J. Electrochem. Soc.*, **146**, 2841 (1999).
8. M. Liu, P. He, J. L. Luo, A. R. Sanger, and K. T. Chuang, *J. Power Sources*, **94**, 20 (2001).
9. P. He, M. Liu, J. L. Luo, A. R. Sanger, and K. T. Chuang, *J. Electrochem. Soc.*, **149**, A808 (2002).
10. G. L. Wei, M. Liu, J. L. Luo, A. R. Sanger, and K. T. Chuang, *J. Electrochem. Soc.*, **150**, A463 (2003).
11. K. T. Chuang, J. L. Luo, G. Wei, and A. R. Sanger, U.S. Pat. pending (2002).
12. E. A. Ticianelli, C. R. Derouin, A. Redondo, and S. Srinivasan, *J. Electrochem. Soc.*, **135**, 2209 (1988).
13. M. Liu, P. He, G. Wei, J. Luo, A. R. Sanger, and K. T. Chuang, Paper 153 presented at The Electrochemical Society Meeting, Philadelphia, PA, May 12-17, 2002.
14. D. S. Thakur, P. Grange, and B. Delmon, *J. Catal.*, **91**, 318 (1985).
15. M. Liu, G. L. Wei, J. L. Luo, A. R. Sanger, and K. T. Chuang, *J. Electrochem. Soc.*, **150**, A1025 (2003).
16. *Dictionary of Inorganic Compounds*, J. E. Macintyre, Editor, 1st ed., Vol. 1, p. 14, Chapman & Hall, London (1992).
17. M. J. Jorgensen and M. Mogensen, *J. Electrochem. Soc.*, **148**, A433 (2001), and references therein.