Short communication

# High performance solid oxide fuel cell operating on dry gasified coal ☆

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## ARTICLE INFO

## ABSTRACT

A fluidized coal bed-solid oxide fuel cell (FB-SOFC) arrangement is employed for efficient conversion of dry gasified coal into electricity at 850 °C. It consists of an anode-supported tubular solid oxide fuel cell of 24 cm² active area coupled to a Boudouard gasifier. A minimally fluidized bed of low sulfur (0.15 wt%) Alaska coal is gasified at 930 °C by flowing $CO_2$ to generate CO. The resulting CO fuel is oxidized at the Ni/YSZ cermet anode. The highest cell power density achieved is 0.45 W cm$^{-2}$ at 0.64 V with 35.7% electrical conversion efficiency based on CO utilization. This power density is the highest reported in the literature for such systems and corresponds to a total power generation of 10.8 W by this cell. Similarly, 48.4% is the highest conversion efficiency measured at a power density of 0.30 W cm$^{-2}$ and 0.7 V. The open circuit voltages are in good agreement with values expected based on thermodynamic data.

## 1. Introduction

Despite the growing concern over increased rate of green house gas emissions and climate change, the role of coal in global energy mix is expected to remain undiminished since it is the cheapest and most abundant fossil-based primary energy resource on the basis of proven reserves [1]. By all accounts, coal will continue to be used as a major source of energy for several decades [2]. Thus the main question concerns how to use coal efficiently and with minimal environmental impact.

Direct utilization of coal in a fuel cell offers an effective way for efficient conversion of its chemical energy into electricity [3]. This objective in the past has met with many practical difficulties

since it is difficult to directly oxidize coal electrochemically. Previous approaches [4] including the more recent FutureGen process [5] that aimed towards the utilization of coal in solid oxide fuel cells (SOFC) have all relied on gasification using steam (reforming). This not only necessitates the need to use a precious natural resource, water, but also makes the process more cumbersome.

A desirable approach is the dry gasification of coal and utilization of CO directly as fuel in a SOFC without the need to use water [6–9]. Indeed, a recent thermodynamic comparison indicated no significant difference in the resulting conversion efficiencies of a fuel cell driven by steam gasification as opposed to dry gasification [3].

Towards this objective, one of the possible approaches includes partial oxidation of coal into CO using air. Unfortunately, this not only dilutes the fuel (due to the presence of nitrogen), but it also leads to NO$_x$ emissions. More importantly, this makes subsequent separation and recovery of $CO_2$ difficult and expensive.

Another possible approach is the use of pure oxygen, which in principle can be obtained by pressure-driven air separation using mixed ionically electronically conducting (MIEC) ceramic membranes [10]. Many efforts on separation of oxygen from air using MIEC membranes are currently underway. These are doped oxide materials of mostly perovskite structure containing transition elements. At elevated temperatures (~800 °C) these oxides can selectively transport oxide ions under an imposed chemical potential gradient in oxygen. An MIEC reactor can be fed on one side with pressurized air. Only oxygen permeates through the membrane thus generating a stream of pure oxygen. This stream can then be

used to partially oxidize coal. Alternatively, MIEC reactor can be directly integrated with a gasifier to form CO by partial oxidation. The reaction is

$$C_{(coal)} + 1/2 O_{2(g)} \rightarrow CO_{(g)} \qquad (i)$$

At 1100 K, the standard Gibbs energy change is $-209\,kJ\,mol^{-1}$ and the enthalpy change is $-112.6\,kJ\,mol^{-1}$. This exothermic heat can be used for maintaining the reactor at temperature. The CO formed can then be fed to a SOFC. The net electrochemical reaction in the SOFC will be

$$CO_{(g)} + 1/2 O_{2(g)} \rightarrow CO_{2(g)} \qquad (ii)$$

The standard Gibbs energy of this reaction is $-187\,kJ\,mol^{-1}$. Thus, the maximum possible (reversible) electrical work with all species in their standard states is $187\,kJ\,mol^{-1}$. Obviously, the SOFC is operated at some finite current and hence, not reversibly. So there are other losses, making the actual electrical work less than this theoretical value.

Combining reactions (i) and (ii), the overall reaction is

$$C_{(coal)} + O_{2(g)} \rightarrow CO_{2(g)} \qquad (I)$$

However, this approach adds significant cost of the MIEC reactor because of the need for pure oxygen. In addition, there also is the requirement of pressurization of air, which means additional equipment and process complexity as well as mechanical and thermal stability of the ceramic membrane under large pressure differentials in addition to operating at high temperatures.

An alternate approach was originally proposed by Gür and coworkers [6–8,11]. The approach consists of using a fluidized bed Boudouard gasifier integrated with a solid oxide fuel cell. This concept is the subject matter for this paper.

In this scheme, depicted in Fig. 1, the coal bed is fluidized using a stream of $CO_2$. The Boudouard gasification reaction is

$$CO_{2(g)} + C_{(coal)} \rightarrow 2CO_{(g)} \qquad (iii)$$

At 1100 K, the standard Gibbs energy change is $-22.194\,kJ\,mol^{-1}$. The enthalpy change is $+168.96\,kJ\,mol^{-1}$ and the entropy change is $174.36\,J\,deg\,mol^{-1}$. Since the process is endothermic heat has to be supplied. The CO that is supplied from
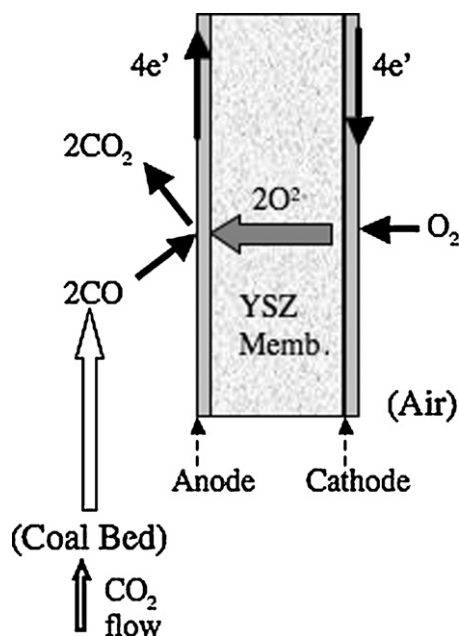


**Fig. 1.** Schematic depiction of the operating principle of the Boudouard gasifier/SOFC couple.

the Boudouard gasifier is electrochemically oxidized in the SOFC. The reaction is

$$2CO_{(g)} + O_{2(g)} \rightarrow 2CO_{2(g)} \qquad (iv)$$

At 1100 K, the standard Gibbs energy change is $-373.72\,kJ\,mol^{-1}$, corresponding to a cell OCV of 0.968 V. Thus, of the two moles of $CO_2$ produced by reaction (iv) in the SOFC, one is fed back to the gasifier. The remaining one mole of $CO_2$ is collected for sequestration or any other possible use. Part of the electrical work derived in reaction (iv) is used up in heating the gasifier (endothermic process). Thus, the net (reversible only) work derived in the process is $\sim 205\,kJ\,mol^{-1}$. Combining reactions (iii) and (iv), the net reaction of the overall process is again,

$$C_{(coal)} + O_{2(g)} \rightarrow CO_{2(g)} \qquad (I)$$

Based on the maximum possible work (i.e., reversible work) that can be derived, the latter process is slightly superior. Since no MIEC reactor is needed nor is it necessary to pressurize air, this process is inherently simpler. Moreover, while the transport of oxygen through the MIEC does not result in useful work, oxygen transport through the electrolyte membrane in the SOFC is primarily responsible for the generation of electrical work. For the reasons discussed above concerning cost, efficiency and minimal complexity, the preliminary conclusion is that the approach using a Boudouard gasifier is superior. Hence, the objective of the present work is to investigate the operation of an SOFC on CO obtained by dry gasification of coal by the Boudouard gasifier.

## 2. Experimental aspects

The solid oxide fuel cells used in this work were of tubular geometry developed by Materials and Systems Research, Inc. The tubular SOFC employed an anode support made of porous Ni/YSZ cermet with $\sim 8$–$10\,\mu m$ thick dense YSZ electrolyte layer and a porous perovskite cathode coating. The nominal dimensions of the cell were as follows: diameter $\sim 1\,cm$ and length $\sim 15\,cm$. The active area (that corresponding to the cathode area) was $\sim 24\,cm^2$. Silver mesh was wrapped around the cathode surface for current collection. The anode current collection was facilitated using a copper mesh inserted inside the tube. Both ends of the cells were open which made it possible to couple the SOFC to the Boudouard gasifier. The gasifier was made of a stainless steel shell 7.5 cm in diameter and 30 cm in length.

The solid fuel employed in this work was an Alaska coal obtained from DOE Coal Sample Bank at Penn State University [12]. It is a Waterfall Seam Kenai-Cook Inlet County, AK, coal with a net calorific value of $28.208\,MJ\,kg^{-1}$ on dry basis, 0.15% sulfur content, 0.975 atomic H/C (Parr) and 0.227 atomic O/C (Parr) ratios. The coal was crushed with the majority of the mass having an approximately 1–3 mm as the particle size with some fines. The coal was pre-treated at 950 °C in nitrogen for 4.5 h to remove volatile matter and produce char.

Approximately 300 g of pre-treated coal char was placed inside the gasifier. Pure $CO_2$ was preheated to 750 °C and passed through the Boudouard gasifier at a flow rate of $0.25$–$0.5\,l\,min^{-1}$. The CO formed in the gasifier was passed through the SOFC, maintained at 850 °C. Fig. 2 shows a schematic of the experimental setup depicting the coupled Boudouard gasifier-SOFC operation. The gasifier and SOFC components were provided with individual furnaces for external heating.

The batch system comprising the Boudouard gasifier and the SOFC was operated under various experimental conditions to achieve two objectives: (a) to establish gasifier performance and behavior. (b) To determine the performance of the integrated Boudouard gasifier-SOFC system. A test stand designed and constructed in-house equipped with automated mass flow controllers
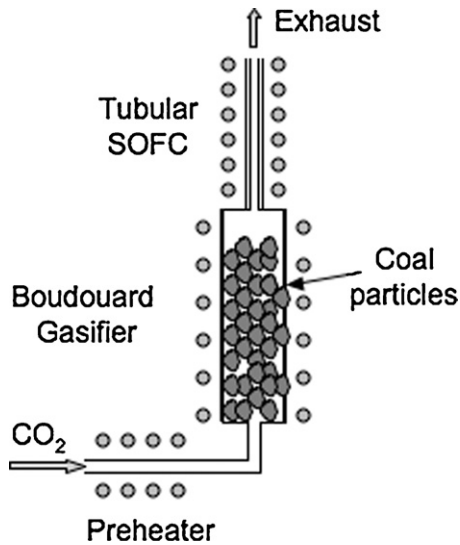
**Fig. 2.** A schematic of the experimental setup showing the fluidized bed Boudouard gasifier coupled to a tubular SOFC.

for gas mixing, an automated electronic load box and power supply for SOFC performance testing, instrumentation for temperature measurement, a gas chromatograph (Varian 3400) for gas analysis, and data acquisition for control and monitoring of experiments was used for this study. Testing was done over a range of conditions such as fuel composition, fuel utilization, and flow rates.

## 3. Results and discussion

Initial tests were conducted using bottled CO in order to establish baseline behavior and performance of the tubular SOFC employed in this work. These experiments were performed using pure CO as well as $CO/CO_2$ mixtures supplied from gas cylinders directly into the fuel cell without any solid fuel present inside the gasifier. Measured open circuit voltage (OCV) values for CO and $CO/CO_2$ mixtures were consistently in general agreement with the theoretically expected values. The voltage–current–power density (V–I–P) behavior of a typical cell at 850 °C is shown in Fig. 3. The open circuit voltage (OCV) of ~1.04 V obtained in pure CO is slightly

lower than the expected value and corresponds to a $CO/CO_2$ ratio of about 14 or a mixture containing ~93% CO and ~7% $CO_2$, effectively. This suggests the possibility of a minor leak through the seals into the cell.

Tests were conducted in pure CO at two different but fixed flow rates that correspond to 60% and 80% fuel utilization at a defined current density. Hence, the results are presented separately for the two flow rates in Fig. 3(a) and (b) and show the cell voltage, power density and cell efficiency as a function of current density. It was assumed that CO oxidation proceeds most likely in a Faradaic manner and the coulombic efficiency of the cell remains close to 100%. The electrical conversion efficiency can then be defined simply by the formula, $\varepsilon = (U_f)(E/OCV)$, where $E$ is the operating voltage of the cell, OCV is the measured open circuit potential, and $U_f$ is fuel utilization based on CO.

For pure CO, the highest performance measured was $0.5\,\mathrm{W\,cm^{-2}}$ at a cell voltage of ~0.64 V at a CO flow rate that corresponds to 60% fuel utilization defined at $1.0\,\mathrm{A\,cm^{-2}}$. The conversion efficiency corresponding to these conditions was 28.8%, while the highest efficiency achieved was 53.3% occurring at a power density of $0.35\,\mathrm{W\,cm^{-2}}$ at 0.7 V using a CO flow rate corresponding to 80% fuel utilization defined at $0.5\,\mathrm{A\,cm^{-2}}$. These results demonstrate that excellent power densities at high efficiencies can be achieved using CO directly as fuel in SOFCs. Based on known thermodynamics of the C–CO–$CO_2$ system, the formation of solid carbon is not expected even when testing at 850 °C as long as the $CO_2$ content is kept above ~5% [13].

Fig. 4 shows performance characteristics of the Boudouard gasifier using low sulfur Alaska coal char. The gasifier temperature was nominally about 930 °C for most experiments. The feed gas was pure $CO_2$. The space velocity of the $CO_2$ feed gas was varied between ~0.005 s$^{-1}$ and ~0.043 s$^{-1}$. The gas mixture exiting the gasifier was analyzed using a gas chromatograph. The mole fraction of CO in the exiting gas mixture was found to vary between ~0.96 for the lowest space velocity to ~0.8 at the highest space velocity used in the experiments. At 900 °C, the equilibrium $CO/CO_2$ ratio in equilibrium with solid carbon at a total pressure of one atmosphere is ~36/1 [13]. The present results thus show that at the low space velocity, equilibrium between CO, $CO_2$ and C was established. At higher space velocities, equilibrium was not established. In concept, it is a straightforward matter to design a gasifier such that the exiting gas is as close to C–CO–$CO_2$ equilibrium as possible, without making the gasifier excessively large.
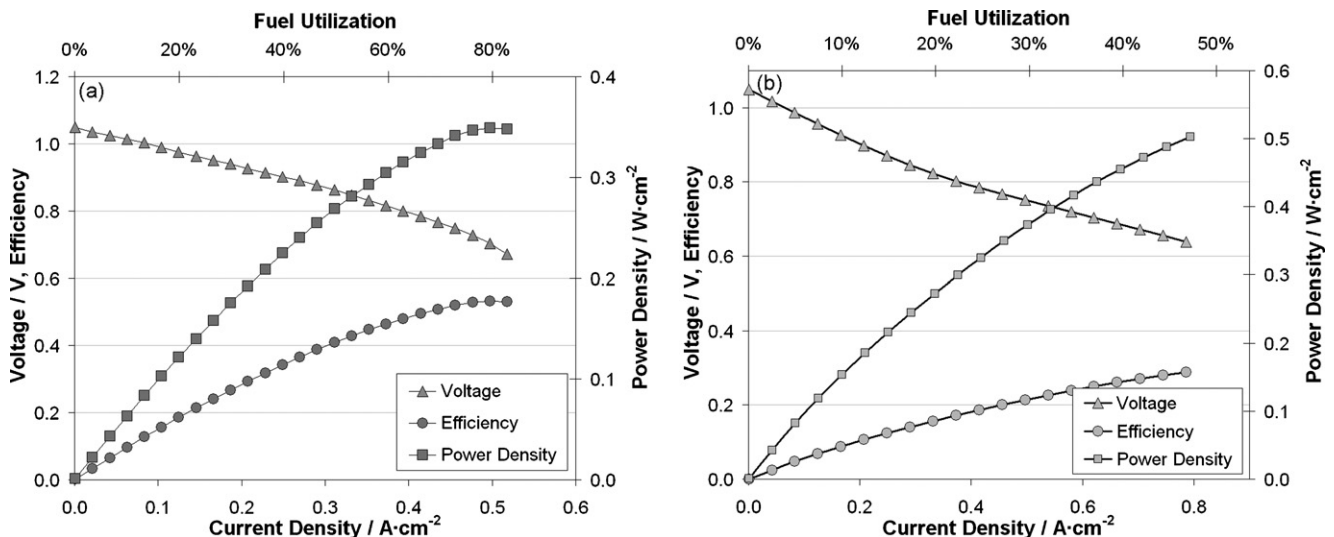


**Fig. 3.** Benchmarking cell performance tests on pure CO at 850 °C for two values of fixed flow rates corresponding to 80% (a) and 60% (b) CO fuel utilization defined for current densities of $0.5\,\mathrm{A\,cm^{-2}}$ and $1\,\mathrm{A\,cm^{-2}}$, respectively.

**Fig. 6.** Performance stability of the SOFC operating at 850 °C directly on gasified Alaska coal char. The current density was maintained at 0.33 A cm$^{-2}$.

Cell performance results on gasified Alaska low sulfur coal are shown in Fig. 5, again for two different effluent flow rates from the Boudouard gasifier that correspond to 60% and 80% CO fuel utilization at a defined current density. The gasifier was connected to the SOFC although no attempts were made for tight physical or thermal integration. The main objective was to test the fundamental concept of operating a SOFC on dry gasified coal using a Boudouard gasifier. The SOFC was maintained at 850 °C and the gasifier was at 936 °C during this experiment.

For the fixed flow rate from the gasifier corresponding to 80% CO utilization at 0.48 A cm$^{-2}$, Fig. 5(a) indicates a peak conversion efficiency of 48.4% based on CO, at a peak power density of 0.30 W cm$^{-2}$ and a cell potential of 0.7 V. This conversion efficiency is in general agreement with the efficiencies predicted in a separate study [3] for fuel cells operating at 0.7 V and employing various solid fuels.

Similarly, at a flow rate corresponding to 60% CO utilization defined at 0.72 A cm$^{-2}$, the electrical conversion efficiency was 35.7% at a power density of 0.45 W cm$^{-2}$ and a cell potential of 0.64 V. To the best of authors' knowledge, this is the first time direct operation on gasified coal with such high values of power density and measured efficiencies has been demonstrated.

Fig. 6 shows the results of performance stability test at a current density of 0.313 A cm$^{-2}$ or about 8 A of total current through the cell at 850 °C. The gasifier used was a batch type and so the coal consumed was not replenished. Over a period of some 30 h, the cell voltage dropped gradually from 0.8 V to ~0.71 V. The initial degradation rate was about 1.3 mV h$^{-1}$. Towards the end of the test, the rate of degradation was greater and the voltage also fluctuated significantly. This decrease in performance and the fluctuations was primarily related to loss of fuel. This assumption was supported by estimates of bed contents. During the 36-h test run, approximately 45% of the carbon in the batch gasifier was consumed. Testing of the gasifier was continued, even after the cell had begun to show degradation, in order to generate a fully depleted ash bed for further analysis. The flow rate of CO in the gasifier



**Fig. 5.** Cell voltage, efficiency, and power density vs. current density of the SOFC operated at 850 °C on gasified Alaska coal char at two values of fixed flow rates corresponding to 80% (a) and 60% (b) CO fuel utilization defined for current densities of 0.48 A cm$^{-2}$ and 0.72 A cm$^{-2}$, respectively.
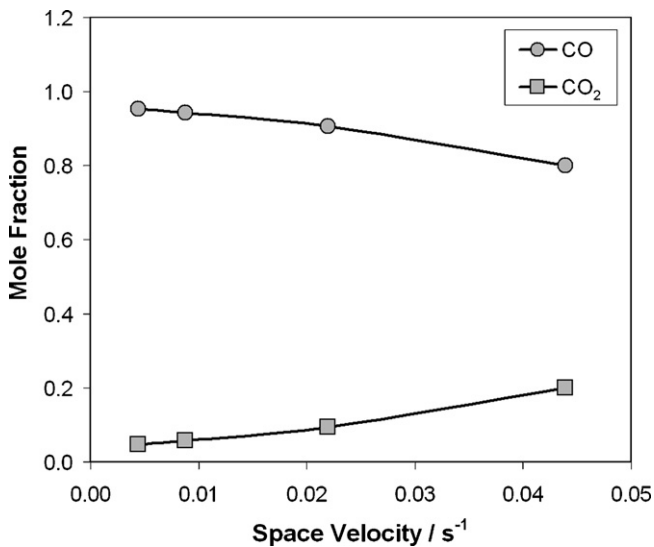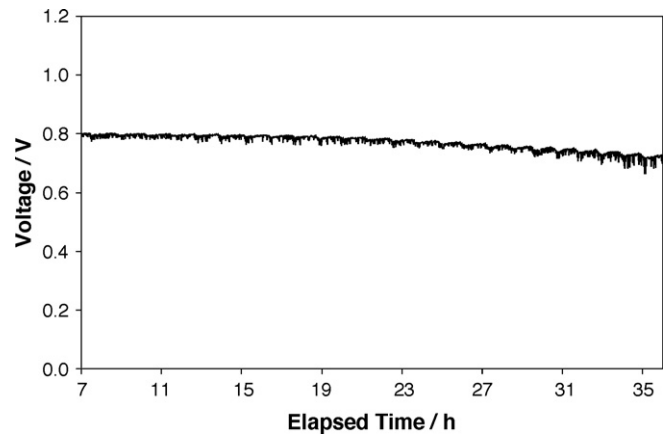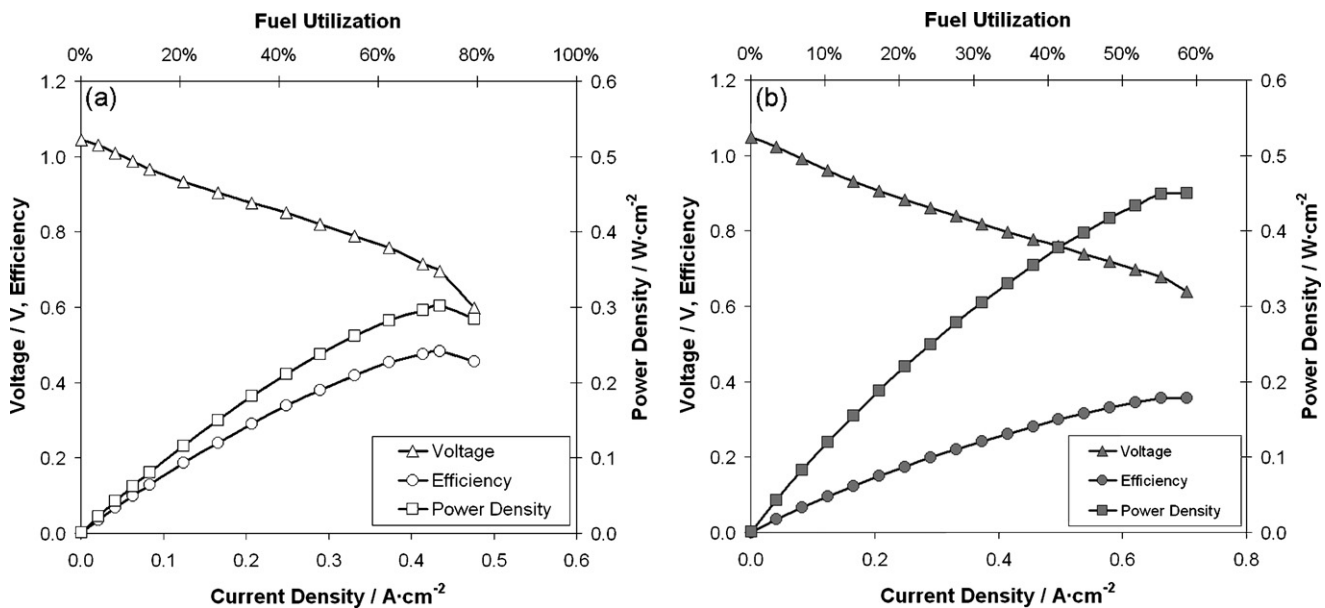
product stream was measured after 60 h under the same flow conditions and it was found that the value had reduced from its initial value by 24%. In other words, as the carbon content of the gasifier was reduced, the $CO/CO_2$ ratio of the mixture exiting the gasifier decreased monotonically, which led to gradual reduction in the cell voltage at constant current.

Interestingly, Fig. 6 indicates no obvious signs of degradation associated with sulfur poisoning over the duration of the test. It is known that degradation due to sulfur normally sets in during the first few hours of operation. Since no gas analysis for sulfurous compounds from the anode chamber was performed, we can only speculate that the absence of obvious signs for sulfur poisoning may in part be due to removal of significant amounts of the inorganic portion of sulfur in the coal, present in the form of sulfides – mostly pyrite – and sulfates via gasification by the evolving hydrogen and moisture in coal as the volatile matter was baked out during the charring process. Since hydrogen (or sulfur) in coal cannot be removed completely by charring, the remaining organically bound sulfur in coal may escape, possibly in the form of COS and $H_2S$, into the anode chamber evidently at insufficient quantities that fall short of triggering the poisoning of the Ni/YSZ cermet anode within the duration of the experiment.

To lend support to this hypothesis, an independent experiment on cell stability was carried out at 850 °C, where controlled and increasing quantities of $H_2S$ were spiked into a 50:50 $CO–CO_2$ fuel mixture. The cell used for this experiment was of the same kind, architecture and composition employed in the present work, with 24 $cm^2$ active cathode area. The results presented in Fig. 7 show that the cell voltage at 0.33 A $cm^{-2}$ is stable below 4 ppm $H_2S$. It starts to show gradual degradation above 4–5 ppm, which seems to accelerate upon increasing the $H_2S$ content to above 9–10 ppm in the $CO–CO_2$ fuel mixture. These independent results suggest that the sulfur release from the Boudouard coal gasifier into the anode chamber of the fuel cell most likely stays below 5 ppm during the duration of these experiments. However, this hypothesis was not confirmed by gas analysis.

After the performance tests, the SOFC anodes were visually examined and no obvious signs of carbon deposition, or coking, were observed. Independent tests in similar cells performed under galvanostatic conditions in flowing pure CO also indicated long-term stability of these anode-supported SOFC cells with no sign of degradation over several hundred hours [14].
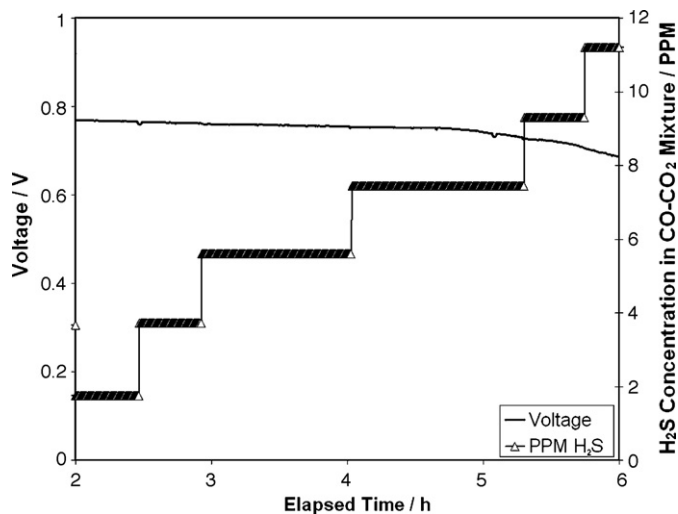


**Fig. 7.** Stability test results of a cell at 850 °C with Ni/YSZ cermet anode running galvanostatically at 0.33 A $cm^{-2}$ and 50% fuel utilization at fixed flow using a 50:50 $CO–CO_2$ fuel mixture spiked stepwise with increasing amounts of $H_2S$.

From a purely thermodynamic point of view, electrochemical oxidation of CO at the anode pulls the gas composition towards higher $CO_2$ compositions, moving it further away from the carbon deposition boundary [15]. Moreover, carbon deposition is not expected at 850 °C above $CO_2$ content of ~5% based on the known thermodynamics of the $C–CO–CO_2$ system [13]. Based on kinetic considerations, carbon deposition can also occur due to the catalytic effect of Ni, which dissolves carbon and redeposits it on its surface upon reaching supersaturation. Indeed, this mechanism is basic to the production of industrial grade diamonds commercially.

However, carbon deposition may still be possible under open circuit conditions, especially if the gasifier temperature is much higher than the SOFC operating temperature. This is because the $C–CO–CO_2$ equilibrium (i.e., Boudouard reaction) shifts to CO formation at higher temperatures. Thus, if the gasifier is at a much higher temperature than the SOFC, the CO disproportionation reaction may occur leading to carbon deposition.

But even if the SOFC temperature is lower than the gasifier temperature, carbon deposition is not expected to accumulate under current carrying conditions when the cell is in operation. If carbon is deposited under OCV conditions, it is expected that during cell operation it will be oxidized. Indeed, a recent report by Pomfret et al. [16] employed in situ Raman spectroscopy to study CO oxidation on Ni/YSZ cermet SOFC anodes at 715 °C and observed direct evidence of graphitic carbon formation under open circuit conditions. However, under current carrying conditions, they reported that the Raman spectra clearly showed the gradual disappearance of the graphitic carbon peak within several minutes. This process was increasingly faster at larger cell overvoltages.

Moreover, in several recent studies it was reported that carbon deposited thermally on SOFC anodes under open circuit conditions from the pyrolysis reaction of methane and propane was oxidized electrochemically to produce power, demonstrating SOFC operation in a rechargeable battery mode with many charge/discharge cycles that involved pyrolytic carbon deposition under OCV and subsequent oxidation of this carbon during SOFC operation [17–19]. These studies provide further support that any carbon deposited on the Ni/YSZ cermet anodes in this study was most likely removed under current carrying conditions by oxidation with the evolving oxygen. The collective results of these recent studies provide strong support that carbon deposition from CO as the primary SOFC fuel, unlike typical hydrocarbon fuels containing hydrogen, should not lead to operational problems with coking under current carrying conditions. This provides another incentive to consider and develop CO-fueled SOFCs.

The total maximum power generated in this work was 10.8 W (at 0.45 W $cm^{-2}$), which is the highest power density and total power reported in peer-reviewed literature for direct utilization of coal for any fuel cell configuration. These results are encouraging and suggest the possibility of practical applications for the FB-SOFC approach towards large scale production of electricity from coal. Naturally, this poses critical questions regarding scale up issues. The tubular cells employed in the present work are essentially the same size (~1 cm OD) as currently being pursued by others for large scale applications using reformed fuel and hydrogen as fuels. Additionally, the present approach is not restricted to tubular design but can be readily extended to planar SOFC. Indeed, we are currently developing a kW scale planar SOFC operating on CO as fuel, and the results of that work will be reported in a separate communication [14].

Although scaling of this process is outside the scope of this work, it is important to note that FB-SOFC offers the flexibility in the stack design to use tubular as well as planar SOFC configurations either in anode-supported or cathode-supported architecture. Moreover,

the FB-SOFC approach presented here is modular in nature, as typical of most electrochemical systems, and lends itself to scaling up the capacity without significant limitations by economies of scale. In addition, further development of this particular approach will greatly benefit from and leverage the advances made in SOFC materials and fabrication as well as scaling up methodologies developed in the DOE-SECA (DOE-Solid State Energy Conversion Alliance) program [20] that aims for MW scale coal syngas-driven IGFC (integrated gasification fuel cell) power plant prototypes. It is therefore reasonable to expect that the advances achieved in this work could be applied at scales suitable for commercial and industrial power generation.

## 4. Conclusions

The present study has shown that coal can be directly utilized in SOFC by dry gasification using the Boudouard reaction. This scheme excludes nitrogen from the process stream and prevents dilution of both the fuel and the product gas $CO_2$ by nitrogen. Accordingly, the resulting exhaust stream of pure $CO_2$ allows for easy capture and recovery of $CO_2$ for possible sequestration. The FB-SOFC approach also does not require or consume water, preserving an increasingly scarce resource. Simple analysis also shows that the use of the Boudouard gasifier may be superior to the more expensive approach using pure oxygen to gasify coal, such as that envisioned using an MIEC reactor. Coupled to a low sulfur content fluidized coal bed, maximum cell power density of $0.45\,W\,cm^{-2}$ was achieved at $0.64\,V$ with $35.7\%$ conversion efficiency and maximum electrical conversion efficiency of $48.4\%$ was demonstrated at $0.30\,W\,cm^{-2}$. Successful operation of an SOFC on dry gasified coal presents significant opportunities for efficient and clean conversion of coal into electricity.

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