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Letter to the Editor

Support for the high efficiency, carbon separation and internal reforming capabilities of solid oxide fuel cell systems

A B S T R A C T

A recent publication by Adams and Barton [1] in the Journal of Power Sources has appropriately espoused and analyzed the high efficiency and carbon separation capabilities of solid oxide fuel cells. Unfortunately, the paper also contains misleading statements and analyses regarding the internal reforming capabilities of solid oxide fuel cells. The current letter to the editor addresses this concern and provides insights into appropriate systems design for solid oxide fuel cells that considers their proven internal reforming capabilities.

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It is appropriate and good that the Journal of Power Sources has agreed to publish a paper that describes the high efficiency and carbon separation capabilities of solid oxide fuel cell (SOFC) technology. These features of SOFC technology are indeed impressive and should be fully expounded and analyzed in the literature. The paper entitled “High-efficiency power production from natural gas with carbon capture,” by Adams and Barton [1], however, makes claims that are not justified by the analyses and contains errors that reduce the impact of the publication and call into question the results.

The claim of the authors in the abstract that they have developed and analyzed a “unique” power system is misleading. The authors may be able to use the word novel, but should not claim that their system is unique. Not only have many companies developed SOFC systems that contain a steam reformer that is heat-integrated with solid oxide fuel cells, but many previous publications have analyzed such systems (see, for example [2–9]). The addition of carbon separation downstream from such a system does not make it unique either. Several studies have previously considered this concept (see, for example [10–16]). I suggest considering the integrated steam reforming system a “standard thermally integrated SOFC system,” while the other configurations (that consider CO₂ reforming for example) may be considered “novel.”

One major problem with the analyses of Adams and Barton is the complete dismissal of the internal reforming capabilities of SOFC technology. I agree with the authors that challenges exist with coking in SOFC anode compartments that process hydrocarbon fuels. But a certain amount of internal reforming with appropriate fuel steam content can typically be accomplished today without coking challenges [6,17–23]. In addition, novel SOFC materials sets have been prominently shown to allow SOFC operation on hydrocarbon fuels without coking [24–28].

The claim of the authors that “chemical equilibrium predictions show that, for methane fuels above 400 °C, graphitic carbon exists as a stable, condensed phase” is only true for certain fuel/steam mixtures. Addition of steam to the hydrocarbon fuel, either externally or internally by electrochemical water production reduces the

coking propensity. The internal processing of methane (and other hydrocarbons) readily occurs in an SOFC and the internal reforming processes affect the performance and thermal integration of the system. The authors themselves claim less than 100% methane conversion in the reformer. As a result, the reformat they consider contains some remaining methane that should not be ignored in the analyses. One should not ignore this feature of SOFC technology and should always consider the physics and chemistry associated with internal reforming when analyzing SOFC technology.

Related to the challenge of coking, the authors should note two additional points: (1) SOFC operation on syngas can also lead to coking whether or not it contains hydrocarbons, and (2) the proposed system contains several other components that are also susceptible to coking under the operating conditions proposed. On the first topic, note that the proposed system involves reforming and shifting upstream of the SOFC, which the authors claim avoids carbon deposition in the SOFC. However, the elements contained in the fuel stream remain unchanged, since none of the components contained in the stream are removed and no constituents are added. Therefore, from an equilibrium perspective, the carbon deposition tendency is unaffected. On the second topic, the proposed system configuration and analyses suggest that one can preheat natural gas up to 615 °C in the first heat exchanger (HX1) without coking. This is an error. The authors should check the composition that results from such pre-heating of natural gas in the absence of steam. The equilibrium mixture that results from heating natural gas to such a high-temperature without steam contains a very high level of condensed carbon. Similarly, operating the steam reformer and/or pre-reformer at a steam-to-carbon ratio of 0.5 will also result in carbon deposition challenges in these reactors, which the authors do not consider.

The use of separate high-temperature and low-temperature shift reactors with requisite heat exchange lowers the efficiency of the system compared to one in which the carbon monoxide is allowed to shift within the SOFC anode compartment. While this may be a novel approach, it is certainly not one that is desired or required. Carbon monoxide can be readily oxidized in the anode

compartment of an SOFC, either electrochemically or by water–gas shift reactions. If the amount of steam present is sufficient, which is typically the case since hydrogen electrochemistry produces steam in the anode compartment, then CO can be used directly in the anode compartment without concerns for coking.

The anode compartment reactions that are included in the analyses erroneously do not consider the water–gas shift reaction. The authors only consider carbon monoxide electrochemical oxidation, whereas it is known that water–gas shift reactions readily occur in the typical anode compartment of an SOFC [20,21,29].

The authors propose heat integration through “inter-cooling stages” in the SOFC stacks that “provide a large portion the high-temperature [reformer] heat requirement.” They later state that “up to 81% of the reformer heat load” is provided in this manner. However, the inter-cooling stages are not shown in the system schematic, nor are the pressure losses and heat losses associated with the complex set of plumbing and heat exchangers required to accomplish this inter-cooling strategy considered. This oversight will lead to a significant over-estimate of performance.

When analyzing the effects of steam reforming extent upon system efficiency, the potential synergistic effects of SOFC cooling that the endothermic reformation reactions could accomplish are ignored. Typically the SOFC temperature would be controlled by manipulating air flow and reductions in SOFC air flow would typically coincide with decreased extent of reforming. But, the authors do not take this effect into account and thus reach a conclusion that is counter to those found by other researchers; that is, a higher methane content in the fuel stream and higher levels of internal reforming typically lead to higher overall system efficiency [30–33].

The authors selected an operating pressure of 10 bar. Operation under 10 bar pressurized conditions is not currently practical with modern planar SOFCs, primarily due to challenges with sealing and the control of pressure dynamics, which must be very accurate in order to protect the thin electrolyte layer from damage due to high pressure gradients.

The SOFC voltage that the authors assume (0.69 V) at 10 bar is far too low for simulating modern SOFC performance. Recent performance data reported by several manufacturers participating in the U.S. Department of Energy Solid State Energy Conversion Alliance suggest that operating voltages of 0.8–0.85 V can be achieved at reasonable current density (e.g. 350–500 mA cm⁻²) [23,33,34].

Some additional minor points should be made.

- (1) Natural gas that is fired in current gas turbine combined-cycle plants is not typically desulfurized before combustion as the authors claim in their introduction. Desulfurization is not required for the gas turbine performance or to meet emissions requirements. Desulfurization is only required for fuel cell system operation on natural gas.
- (2) The exhaust of a combined-cycle plant also contains a high concentration of oxygen which cannot be ignored in the CO₂ separation process—the authors suggest that only nitrogen is present in the exhaust.
- (3) Fuel utilization of 92% is too high.
- (4) The most important system design tradeoffs associated with selecting the design operating pressure are the positive improvements in SOFC performance as balanced against the negative parasitic power requirements for pressurization, SOFC sealing challenges and SOFC robustness to pressure differences. The slight decrease in the extent of reformation as pressure increases is nearly inconsequential.

- (5) Temperatures in the anode off-gas reactor will be impractical (too high) if one reacts the stream with a stoichiometric amount of oxygen as the authors suggest in their analyses. In addition, nitrogen-oxide production may result (with only 92% pure oxygen use).
- (6) Assuming a 0 °C approach temperature (pinch point) in the heat exchanger network is not reasonable.

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