



## Response to Letter to the Editor

**Re: “Support for the high efficiency, carbon separation and internal reforming capabilities of solid oxide fuel cell systems”**

We agree with Dr. Brouwer’s enthusiasm in his recent Letter [1] for the enormous potential of SOFCs for electricity production, especially in a carbon-constrained world. We share his hope that this technology will be developed and demonstrated at the large scale. However, we respectfully disagree that the decision to choose internal vs. external reforming for natural gas feeds is settled, particularly for industrial-scale applications which have yet to be demonstrated. Indeed, we feel that internal reforming is promising and valuable, but that external reforming should also continue to receive attention as well, particularly for near-term applications.

We were motivated to choose the external reforming strategy in our recent work [2] because, to advance the technology to the commercial scale, investors must first be willing to risk billions of dollars in “undemonstrated” ventures. In order to overcome this major hurdle, especially for those first few crucial demonstration plants, it makes sense to reduce the risks as much as possible. The three major risks for those first few industrial-scale plants are: (1) demonstration of large-scale SOFCs; (2) demonstration of large-scale SOFCs with fuels other than H<sub>2</sub>; (3) demonstration of carbon capture and sequestration (CCS) technologies. For our purposes, carbon sequestration risks can be avoided by pumping CO<sub>2</sub> to an existing pipeline for enhanced oil recovery, selling CO<sub>2</sub> to another industry, or venting the CO<sub>2</sub> altogether. This leaves the fuel cell risks. There is little doubt that H<sub>2</sub> is a “nicer” fuel for SOFCs than hydrocarbons. H<sub>2</sub> is shown to achieve higher voltages and smoother profiles during operation in recent tests [3] and can be expected to have longer operational lifetimes—hence the need to find solutions such as improved anode materials to handle coking issues from hydrocarbons (which are likewise undemonstrated at the large scale). Therefore, we feel that the easiest pathway toward full-scale development and acceptance of the technology is to focus first on the scale-up issues using the “nicest” fuel possible, even if there are some cost or efficiency penalties. Then, as that major step is completed, the next level of risk can be undertaken, such as using internal reforming with advanced anode materials.

Therefore, we presented a big-picture techno-economic analysis of a large-scale plant with four different methods for external reforming, demonstrating that the economic and environmental rewards are high even at the lowest levels of risk. The objective was not to provide an end-all design, but rather to perform mass and energy balances, address the major implementation issues, and generate cost estimates against standardized baselines (in particular, the NETL’s “Cost and Performance Baseline for Fossil Energy Plants, vol. 1, Bituminous Coal and Natural Gas to Electricity Final Report” [7]). It is our hope that these encouraging results will spur more interest in the technology, incentivize investment and commercialization, and lead to its continuing improvement with other fuels and techniques, including internal reforming. However we

did not “completely disregard” internal reforming either, as it was made clear that it was outside the scope of our analysis, with a representative sample of references mentioned instead.

We now address some of the other points made in the Letter. We did neither intend to claim that the idea of external reforming itself was novel, nor did we intend to claim that the idea of CO<sub>2</sub> capture itself was novel, as we in fact referenced many of the same works and groups throughout our paper as cited in the Letter. The novelty lies in the combination of external reforming, external water gas shift (which is cited as novel in the Letter), and CO<sub>2</sub> recovery, a combination which did not appear to exist in the literature, to our knowledge. In particular, the method for CO<sub>2</sub> recovery is unique since we do not employ the “simple single condenser” approach which is commonly used. This is the basis for a recent patent application on the concept [4].

Contrary to the claims of the Letter, we explicitly discussed how syngas (i.e., reformed natural gas) can cause carbon deposition in the SOFCs in section 2.3 of our paper, as it was the premise for including the water gas shift (WGS) reactors. Additionally, we agree with the author of the Letter that including the lower temperature, exothermic WGS reactions may result in a lower theoretical electrical efficiency, all else being equal. However, the minor efficiency difference will be a small price to pay to reduce the risk of carbon deposition and thus the financial risks of implementation, especially since we demonstrate that the efficiency of the system is still high and electricity costs are low.

The heat integration considered in this paper was not intended to be a definitive, final design. Rather, we looked at theoretical HENs in order to rule out impossible design configurations based on the availability of heat at certain temperatures. For example, for a system relying on heat integration of the SOFCs and reformer, we determined theoretical bounds on the fuel utilization in the SOFCs. Using a simple pinch analysis with a  $\Delta T$  of 0 °C enables the use of fast algorithms to generate these idealized bounds.

Likewise, heat integration between the SOFCs and other parts of the plant is considered in the theoretical sense. Contrary to the claims of the Letter, we do in fact assume a 0.2 bar pressure drop per SOFC inter-cooling stage, or about 1.4 bar across the whole SOFC section (note streams 11 and 12 in Table 1). Additionally, the use of inter-cooling stages has been proposed in other works as well [5]. Furthermore, we do actually consider heat losses from the SOFC for the high temperature heat transfer, assuming that 5% of the free energy of the electrochemical oxidation reactions is lost as waste heat. This is a conservative estimate based on Kuramochi et al. [9], and without which our predicted efficiencies would be even higher. However, we only mentioned this detail implicitly by reference to a parallel work which describes our models in more detail and is currently in press [10]. We regret not calling attention to this detail in the text of our work.

As described in our paper, it may be possible to use the scheme for heat integration between the SOFCs described by Riensche et

al. [8], in which a combined reformer and SOFC is designed such that reforming zones alternate with cell zones, so that heat is transferred directly from the SOFC zones to the adjacent reforming zones by radiation. In their scheme, they report that there is enough heat to reform 75% of the methane, while in our paper, we calculate that we have enough heat for about 80% of the methane (that is 81% of the ~99% of methane converted), which is in very good agreement. While our model mimics a more traditional heat integration approach, it is still a very good approximation for the envisioned integrated system and will have a negligible effect on the results.

In the Letter, criticism is drawn toward the choice of system pressure and voltage, simultaneously arguing that 10 bar is not practical and that our assumed voltage is too low according to manufacturer data at those pressures. First, our assumed voltage (0.69 V) was intentionally conservative. Given the high uncertainty in the actual attainable average voltage at the time of large-scale implementation, and particularly since cells tend to degrade with use, it makes sense to use a more conservative value. We agree with the author of the Letter that this may have underestimated the potential of our system, but this was intentional. Again, in the context of the financial risk in constructing such a plant, we preferred to examine the performance and economics under conservative conditions and show that even then, conditions are very favorable. In our parallel work [10], we quantify the sensitivity of the plant efficiency to changes in the assumed SOFC voltage (though for a coal-based system). To increase the voltage to 0.8 V would give about a 2–3 percentage point increase in efficiency.

With regard to the concerns about pressure, we have three comments. First, our model of the SOFC system applies equally to either planar or tubular SOFCs, and so one could simply use the tubular version if 10 bar planar systems are not yet stable by the time construction of such a facility is scheduled. This is particularly true if one intends to heat integrating the steam reformer with the SOFCs as discussed above. Second, the author of the Letter is correct in stating that the primary system tradeoffs for pressure are the SOFC efficiency improvements vs. the costs of pressurizing the fuel, but this is only true for processes without CO<sub>2</sub> capture. Once CO<sub>2</sub> capture is considered, there is an extra incentive to have high pressures in the SOFC such that the CO<sub>2</sub> in the exhaust can also be recovered at high pressure with an appropriate method such as the CO<sub>2</sub> recovery method we have proposed. This reduces the significant parasitic load necessary to compress CO<sub>2</sub> to supercritical conditions suitable for pipeline transport and sequestration, and impacts the efficiency of the process as a whole as shown in Figs. 2–4 of our paper. Third, all of our design cases maintain equal inlet pressures on the anode and cathode sides, such that the pressure difference between the two is minimized, mitigating the risk of blowout as much as possible.

On the lesser points: though the author rightfully mentions the issue of coking in the natural gas pre-heater (HX1), it is nevertheless irrelevant to the main point. Stream 35 can be diverted into HX1 instead of the pre-reformer, or the temperature of HX1 can be appropriately reduced, either of which will have a small efficiency penalty. However, for theoretical purposes, our approach is commonly used in other works [5,6].

We take advantage of a relatively low O<sub>2</sub> purity from the ASU (92%) because it is cheaper to generate than high-purity O<sub>2</sub>, but still pure enough to ensure that the amount of N<sub>2</sub> in the CO<sub>2</sub> pipeline remains within specifications. Higher purity O<sub>2</sub> will invoke higher costs, but result in a higher purity CO<sub>2</sub> product, which may or may

not be important depending on the circumstances. Moreover, NO<sub>x</sub> formation is less of an issue, since any NO<sub>x</sub> produced will ultimately end up in the CO<sub>2</sub> pipeline destined for sequestration, rather than the atmosphere. The final choice of the O<sub>2</sub> purity will be a detailed design decision.

We agree with the author that NGCC flue exhaust contains O<sub>2</sub> from the use of excess air, and we regret that our wording may ambiguously imply that only nitrogen was present. It is common to use the CO<sub>2</sub>/N<sub>2</sub> separation problem as the simplest way to describe the challenges of post-combustion capture of CO<sub>2</sub>, prompting the need for alternative strategies (such as IGCC for coal, oxy-combustion, chemical looping combustion, etc.). Our report referenced the NETL's study [7] for NGCC with CCS as the baseline case, which included both O<sub>2</sub> and N<sub>2</sub> in the relevant calculations.

Finally, the claim that we did not consider carbon deposition issues in the pre-reformer and reformer is also incorrect. As stated in section 2.2, the pre-reformer steam level was set at H<sub>2</sub>O/C = 0.5 (where C is for all carbon present, not just the higher hydrocarbons), which is in the range of ratios deemed necessary and sufficient for conversion of higher hydrocarbons without carbon deposition according to the two references cited in that paragraph. If desired, higher ratios can be achieved rather trivially by diverting a portion of stream 36 to the pre-reformer, at the price of requiring a higher volume pre-reformer. The reforming section uses much higher ratios; for example, the case in Table 1 uses a ratio of H<sub>2</sub>O/C = 6.7. Figs. 2–5 highlight how the efficiencies of the plant would change if one desired to add additional steam to the reformer for the various design scenarios.

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