Perspective

Low-Carbon Electricity Is Great: What about “Less-Carbon”?
Turgut M. Gür*†‡
Department of Materials Science and Engineering, Stanford University, Stanford, California 94305, USA

This article posits a technological roadmap for the continued use of fossil fuels for power generation with total carbon capture. Combustion-based power from fossil fuels carries great risks for the sustainability of our planet. Less-carbon power generation in high temperature fuel cells provides the best option for achieving sustainable use of fossil fuels, albeit not without their own challenges. They offer efficient conversion with near capture-ready CO2 emissions. They eliminate significant energy penalties and costs associated with carbon capture, and also diminish the need for freshwater withdrawal. Unlike low-carbon technologies that refer to the carbon content or the generation source on the input side of power generation, "less-carbon" concept relates to the output side of generation, and specifically demands reduced emissions with total carbon capture.

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As atmospheric CO2 concentration now exceeds 400 ppmv, the 21st century poses conflicting demands on sustainability for our carbon-constrained planet. The critical question is how to meet the world’s ever increasing demand for electricity generation, while reducing CO2 emissions by more than 50% by mid-century. Limiting the global temperature rise to below 2 °C with reference to the preindustrial level requires the energy sector to become carbon neutral by 2100.

Shifting electricity generation to low-carbon sources such as renewables, biomass, and even natural gas are critical steps for transitioning toward a sustainable energy economy. However, low-carbon electricity does not necessarily require capturing of the emitted CO2, but rather refers to the carbon content of the fuel source or the generation technology. In other words, it relates primarily to the input side of power generation. Instead, this article introduces the concept of less-carbon electricity that relates specifically to the output side of generation, and explicitly requires significant reduction in CO2 emissions produced in capture-ready form.

According to the U.S. Department of Energy’s (DOE) International Energy Outlook 2016 report, world’s electricity generation is expected to grow by 69% to 36.5 trillion kWh in 2040, where natural gas and renewables will be the fastest growing sectors, the latter at 2.6% per year. It also forecasts coal to remain the single largest resource for electricity generation into the foreseeable future. The shares of solar, wind, and nuclear in electricity generation in 2040 are expected to be, respectively, 2.6%, 6.7% and 12.3%, while coal (29.1%) and natural gas (27.8%) will continue to dominate power production with a combined share of nearly 60%. Clearly, it is unrealistic to wish away or dismiss fossil fuels just yet.

With recent advances in wind and solar, their cost of electricity now seems to compete favorably with coal-fired power in some places. However, the prospects for intermittent renewables to dominate utility scale power generation are still limited by lack of proven, cost effective, and scalable electricity storage technologies. This handicap arguably remains to be the “ball and chain” of solar and wind. Indeed due to insufficient storage, Bloomberg Technology News reported on July 31, 2017 that California had to forego more than 300,000 MWh of solar and wind power in the first half of 2017, while Germany tossed out 4% of wind energy in 2015 and China has discarded 17% of its renewable energy.

Indeed, the global electricity storage capacity is only around 145 GW of which pumped hydro provides the lion’s share (132 GW in 2012), while the combined installed capacities for electrochemical and mechanical storage technologies were merely 1.5 GW in 2014. Currently, Tesla has installed only 300 MW of battery storage capacity, and AES and Siemens have battery storage projects in 13 countries with a combined capacity of 463 MW. Obviously, these numbers fall far too short of the current storage demands for the global installed capacities of solar (234 GW) and wind (460 GW). The 2015 Paris Agreement aims at achieving 37% renewables by 2040, while 60% is needed for carbon neutrality by 2100. These are ambitious challenges to meet. Accordingly, banking the future of electricity generation entirely on intermittent renewables seems unrealistic.

Current Status

Historically, fossil fuels have been the backbone of power generation and currently provide nearly 2/3rd of the world’s electricity, while coal alone is responsible for 40%. However, for every kWh of electrical energy generated, coal-fired plants spew about 1000 g of CO2 into the environment, while natural gas plants emit nearly 470 g of CO2 per kWh. Clearly, this is not sustainable. In recent years, coal-based power in western countries has decreased in favor of natural gas. However, this is somewhat deceptive, as CO2 emissions do not recognize national or continental boundaries. While coal demand in North America and Europe has indeed decreased nearly 50% between 2000 and 2015, demand for coal in Asia has increased by 60% within the same period. In fact, coal’s share in electricity generation in China and India hovers well above 70%. Even in Denmark and Germany, which have invested heavily in renewables, coal-based power accounted for more than 40% of their electricity production in 2013. Globally, atmospheric emissions monotonically rose at a rate of ~2 ppmv/yr to nearly 39 GtCO2/yr in 2014.

For sustainability, we must design a different future for fossil fuel-based power generation. Burning them is unsustainable and too risky. Hence, less-carbon technologies are needed that also allow total carbon capture. Yet, a large fraction of coal and natural gas power plants across the globe still operate using the century-old technology of combustion, where typically the fossil fuel is burned in air, and the thermal energy of combustion is first converted into mechanical energy in a turbine, and finally to electrical energy in a generator. This three-step process chain is energetically wasteful and yields efficiencies in the low 30% for combustion-based coal and natural gas fired power generation.

Conversion in high temperature fuel cells potentially increases conversion efficiencies with total carbon capture, and offers the best option for “less-carbon” power with significantly smaller carbon-footprint. As a critically important distinction, this electrochemical conversion process involves not combustion but oxidation. High exothermicity of the oxidation reactions also produces high quality waste heat that can be used in a bottoming cycle to add 5–10 percentage points to the overall efficiency. Also, nitrogen is excluded from the cell proper and does not enter the process stream. This has two important consequences. First, no post operation for CO2 separation is necessary, which is an expensive and energy intensive process. Similarly, no air separation unit (ASU) for oxygen production is necessary, which is otherwise required for oxy-combustion, integrated
gasification combined cycle (IGCC), and natural gas combined cycle (NGCC) technologies.

To improve efficiency, solid oxide fuel cells (SOFC) have recently been considered for incorporation into the downstream of the external gasification and reforming steps of IGCC and NGCC processes, and are called, integrated gasification fuel cell (IGFC) - for coal, and natural gas fuel cell (NGFC), respectively. DOE’s Advanced Energy Systems (formerly SECA) program, as well as Bloom Energy’s commercial SOFC power plants are based on this scheme.

In contrast to IGFC and NGFC, physical and thermal integration of gasification or reforming steps inside the anode compartment of the SOFC potentially offers still higher conversion efficiencies, with the possibility of direct electro-oxidation of the fuel. Recent progress in coal and natural gas conversion via internal gasification or reforming inside high temperature fuel cells has been reviewed elsewhere. Indeed, advances in electrocatalytic oxidation of methane in SOFCs have demonstrated great promise for efficient conversion of natural gas to electricity with total carbon capture. Similarly, recent progress resulted in practically significant power densities for the conversion of solid carbonaceous fuels, including 878 mW/cm² reported at 750 °C using a waste-derived pyrolyzed carbon in a hybrid molten-carbonate/solid oxide electrolyte fuel cell configuration, and 450 mW/cm² reported at 850 °C for untreated Alaska coal in a solid oxide-based fuel cell.

Fuel cells for fossil fuel conversion explored different types of cell configurations. Cells using molten electrolytes cannot convert natural gas directly and usually pose stability problems for cell components because the electrolytes, such as molten carbonates or hydroxides, are typically corrosive in nature and prone to parasitic reactions. Nevertheless, conversion of pyrolytic carbon in molten carbonate cells demonstrated exceptional efficiencies approaching 80%, albeit with sluggish reaction and transport rates typical of molten/solid systems. Conversely, fuel cells based on solid oxide electrolytes offer faster electrode kinetics and simpler operation typical of gas/solid systems, and can convert both solid and gaseous fossil fuels via internal reforming or gasification.

Conceptual schematics of less-carbon conversion of fossil fuels in solid oxide-based fuel cells are presented in Figure 1, where the net electrode reactions as well as the respective reaction pathways are illustrated with arrows. The water-gas shift reaction, although active at these temperatures, are omitted from the figures for simplicity.

The primary outputs are electricity and a mixture of steam and CO₂ with small amounts of unconverted CO in the exhaust stream. Upon condensation, CO₂ is easily captured.

Moreover, no external water input is needed for the internal gasification or reforming reactions at the anode chambers, which is important for water security. As all fossil fuels contain hydrogen in their chemical makeup, positive feedback loops for CO and H₂ formation are initiated as shown by the arrows in Figure 1. These shuttle mechanisms continuously supply CO and H₂ to the anode surface, where the oxidation products CO₂ and H₂O are internally consumed in subsequent gasification and reformation reactions in the anode chamber. By comparison, up to 1200 gallons of raw water are required for every MWh of electricity for various coal and natural gas-based power generation technologies. Carbon capture and storage (CCS) further increases raw water usage by up to 90% for coal-based and 96% for natural gas-based power technologies.

The theoretical efficiency for direct electrochemical conversion of carbon or methane in fuel cells is 100%, due to the negligible entropy change during their corresponding oxidation reactions. This exceptional theoretical ceiling makes conversion in fuel cells quite attractive, and inherently efficient. U.S. Department of Energy estimates that the practical efficiencies including the corresponding energy penalties for CCS of fossil fuel conversion in fuel cells that are externally coupled to gasification (for coal) or reforming (for natural gas) cycles can exceed 54% for coal in integrated gasification fuel cell (IGFC), and 64% for natural gas fuel cell (NGFC) processes. Moreover, the estimated costs of electricity (COE) including CCS for IGFC ($78.8/MWh) and NGFC ($68.7/MWh) are significantly cheaper than their counterparts without the fuel cell, and further cheaper than for combustion-based power generation (> $130/MWh).

Recently, SOFC power plants running on externally reformed natural gas have been commercialized by Bloom Energy of California, in capacities up to 250 kW. These SOFC power plants are rated at 50–65% efficiency based on the lower heating value (LHV) of natural gas. Naturally, direct electro-oxidation of methane as opposed to external reforming is expected to result in even higher conversion efficiencies.

Collecting capture-ready CO₂ directly from the fuel cell exhaust is critically important not only for environmental sustainability, but also for economic and energetic reasons. CO₂ capture adversely impacts plant efficiency, raw water usage and cost of electricity in a...
significant manner. It constitutes 50–90% of the overall chain cost of CCS.\textsuperscript{29–32} Technologically, the most advanced commercial option for CO\textsubscript{2} capture from N\textsubscript{2}-rich flue streams is chemical separation in alkyl alkanolamine solutions such as monoethanolamine (MEA). However, this benchmark process is costly and energy intensive. The minimum energies theoretically required to capture CO\textsubscript{2} from coal and natural gas combustion plant streams are 31–44 kWh/tON\textsubscript{2} and 38–57 kWh/tON\textsubscript{2}, respectively.\textsuperscript{9,52} Practical energy penalties for CO\textsubscript{2} capture are significantly higher. Indeed, energy penalty for CO\textsubscript{2} separation can demand 25–40% of the energy produced at the power plant and make up 70% of the additional cost for CCS.\textsuperscript{31} A similar study estimated the energy penalty for CO\textsubscript{2} capture to be 50–80% of the power produced from coal-fired plant, and around 40% for IGCC and NGCC.\textsuperscript{34} The financial cost of CO\textsubscript{2} capture is estimated to be nearly $125/tON\textsubscript{2} for natural gas plants, and $60–70/tON\textsubscript{2} for new and $73–107/tON\textsubscript{2} for old coal-fired plants.\textsuperscript{34}

Another energy penalty for CCS relates to the ASU unit, as oxy-combustion, IGCC and NGCC all require oxygen separation to produce a capture-ready CO\textsubscript{2} flue stream. Typically, cryogenic ASU consumes 10–15% of the power produced at the plant. Clearly, fossil fuel conversion in fuel cells eliminates these significant costs and additive energy penalties.

Interestingly, high temperature electrolyzers and electrochemical reactors, similar in architecture to SOFCs but with specialized catalytic electrolytes can be employed to electrolyze chemical fuels such as hydrogen,\textsuperscript{35} ammonia,\textsuperscript{36} and/or hydrocarbons,\textsuperscript{53,54} from the oxidation products CO\textsubscript{2} and/or H\textsubscript{2}O by using off-peak or renewable power. Such large-scale storage options in the form of chemicals will help expand the deployment of intermittent renewable sources.

Moreover, high temperature fuel cells, in particular SOFCs, offer fuel flexibility and distributed generation options. Locally available solid or municipal waste, biomass, forestry and agricultural waste products are potential feedstocks for distributed generation of electricity in an environmentally responsible manner. Already, conversion of waste fiberboard,\textsuperscript{33} various biomass,\textsuperscript{39} and plant derived activated carbon\textsuperscript{40} into electricity in high temperature fuel cells have successfully been demonstrated at the laboratory scale.

**Future Needs and Prospects**

Admittedly, conversion in high temperature fuel cells is not without its technical challenges as well as concerns regarding cost, robustness, service life, etc. As the C-H bond is exceptionally strong (435 kJ/mol), methane activation for direct electro-oxidation is difficult to accomplish due to this steep energy barrier.\textsuperscript{41} For this reason, most fuel cell systems employ external reforming of natural gas to circumvent this difficulty. However, recent progress in the development of effective catalytic anodes based on ceria,\textsuperscript{42} doped perovskites,\textsuperscript{43} and various cermet\textsuperscript{25,44} has successfully demonstrated direct conversion of methane and other hydrocarbons in SOFCs. These developments are particularly important in the face of recent shift toward natural gas as a low-carbon energy source.

Direct electro-oxidation of carbon in SOFCs is also challenging and requires intimate physical contact at the atomic scale between the coal particles and the catalytic anode, which confines the electrochemically active area to discrete solid-to-solid points of contact. Instead, internal gasification or reforming reactions at the anode compartment of the SOFC often dominate the reaction pathways (see arrows in Figure 1). By contrast, molten electrolyte-based fuel cells offer intimate contact between the molten electrolyte and the solid fuel providing possibility for direct electrochemical oxidation, but suffer from parasitic reactions, slow interfacial kinetics, and difficulty in maintaining electrical connectivity among the solid fuel particles dispersed in the molten electrolyte.\textsuperscript{22} Thermal and chemical stability of cell materials and components may be mitigated by lowering the operating temperature.\textsuperscript{22} Cell degradation and the adverse impact of fuel contaminants especially by sulfur pose further challenges. Recent advances in understanding sulfur poisoning in SOFCs\textsuperscript{15,46} as well as progress in developing perovskite-based catalytic anodes with enhanced kinetics and redox stability\textsuperscript{47,48} and with sulfur tolerance\textsuperscript{49} gave promising results. Admittedly, cost and lifetime of cell stacks also present concern.\textsuperscript{50} but recent progress by U.S. DOE’s Advanced Energy Systems program foresees reduction in cell degradation rates and respective cost targets for SOFC stacks and power modules to $<225/kW and $900/kW, respectively.\textsuperscript{50} Increasing the power density by 4-fold may bring the stack cost to below $50/kW.\textsuperscript{51} These estimates would compete favorably with other power generation systems.

Admittedly, some of the technical challenges such as catalyst deactivation and cell degradation due primarily to sulfur and other contaminants that are omnipresent in fossil fuels are difficult hurdles to resolve. However, none of the challenges facing fossil fuel conversion in fuel cells seem to be fundamental in nature.

Use of theoretical computational tools can help provide valuable insights and opportunities in materials design,\textsuperscript{52} and surface engineering\textsuperscript{52} including strain effects,\textsuperscript{53} as well as understanding the energetics of catalytic processes at the atomic scale.\textsuperscript{54,55} Recent progress in observing materials properties and rate processes using in situ and in operando analytical and spectroscopic tools promise significant advances in understanding materials behavior for improved performance.\textsuperscript{56} Collectively, these tools can offer insights to help overcome some of these limitations.

**Conclusions**

The technological hurdles along the roadmap proposed in this article is too risky for industry to take on these challenges alone, unless there are formal programs, partnerships or incentives supported by federal governments. A widely executable price placed on carbon emissions will also provide an impetus for rapid progress and help incentivize the industrial sector. As the window of opportunity to mitigate climate change is rapidly narrowing, delaying development of less-carbon generation technologies comes at great risks. Although fuel cells are generally not the panacea for all the ills of fossil fuels, it is the view of this article that the prospects for continued and environmentally responsible utilization of fossil fuels for sustainable electricity generation rests with efficient conversion in high temperature fuel cells that offer reduced emissions with total carbon capture. It is also the intent of this article to stimulate and engage the scientific research community in this important area.

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