TIP White Paper

Electrochemical Conversion of Carbon to Electricity to Reduce Fossil Fuel Consumption and CO_2 Emissions

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Abstract

The finite fossil fuel resources of the world are being consumed at an increasing rate as population and the average standard of living both increase. World oil production is expected to peak within a few years, natural gas production is predicted to peak in about 2050, as is coal production in about 2100. Atmospheric CO_2 concentrations continue to increase beyond current levels of about 350 ppm as a result of fossil fuel consumption. There is general acceptance of the threat of significant global warming unless CO_2 levels in the atmosphere can be stabilized at acceptable levels thought to be about 550 ppm sometime in the future.

Increasing the fraction of the electricity component of total energy consumption has long been related to increases in GDP and societal standards of living. More of the world's electricity is produced by combustion of fossil fuels to produce heat which is converted into electricity by the Carnot Cycle, Brayton Cycle or a combination of the two, than any other technology. Carbon-rich wastes are produced in large quantities by the food, biofuel, forest product, and wood processing industries. Municipal solid waste is another source of carbon-rich fuels. Combustion technology is currently used to convert a small portion of these waste materials to electricity Solid carbon-rich fuels can be converted to electricity more efficiently if combustion technology is replaced with much more efficient electrochemical technology. Typical solid fuel combustion-based power generation systems processes average about 35% today, with new large central station coal plants achieving efficiencies in the 40-45% range and smaller biomass fueled plants averaging about 25%. Electrochemical conversion of solid carbon-rich fuels has the potential to achieve overall system efficiencies of 55-65%.

The predicted high efficiency of electrochemical carbon conversion has been validated experimentally at the single Direct Carbon Fuel Cell (DCFC) level within the range of 70-80%, but significant engineering issues have been identified in the limited R&D small lab-scale work. Significant engineering challenges have been identified in considering different approaches to scale-up to commercial systems, but remain unresolved.

The key point of this paper is that only electrochemical conversion of carbon-rich fuels in DCFC systems has the potential to achieve 60-65% efficiency with 100% CO_2 capture at a Cost of Electricity (COE) and with an estimated capital investment requirement that is lower than any of the conventional or developmental alternatives.

Consideration of the challenges that must be overcome leads to the conclusion that long-term, well funded development programs will be required to achieve commercialization. As a result, private sector investors have been deterred by the anticipated long-time horizon required to achieve a reasonable return on their R&D investments. Federal government funding is needed to accelerate the further development of this promising electrochemical technology through the modular component demonstration phase so that the private sector will have an acceptable basis to invest in further scale-up to economically viable commercial-scale systems.

Keywords: fossil fuel, renewable energy, distributed power generation, direct carbon fuel cell

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1. Introduction

The United States has long been the world largest consumer of energy resources and emitter of the largest amount of CO_2. Our economy requires about 20 million barrels per day of oil, 60% of which is imported, and 3 million tons per day of coal. World oil production is peaking at this time and prices have become very volatile. The public has become increasingly aware of the economic impact of high oil prices on their lives. There is also growing awareness that CO_2 emissions have to be reduced to prevent serious impacts of global warming. Large scale demonstrations of CO_2 sequestration technology, primarily in deep saline reservoirs, are being planned by US DOE. The long-term solution to a world without fossil fuels is seen as a combination of using nuclear energy and renewable energy including biofuels to produce electricity. It implies the use of electricity as an important substitute transportation fuel. In the nearer term, one of the most important strategies to meeting these challenges is to develop and utilize technologies that are more efficient in converting the chemical energy in fuels into consumer usable energy forms such as electricity so that both fuel consumption and CO_2 emissions can be reduced simultaneously.

2. Critical National Needs and Associated Societal Challenges

Table 1 shows the current use of energy resources for world and US energy consumption. Oil use in the US is driven primarily by the need for transportation fuels. Coal, nuclear, and renewable fuels are used almost exclusively for electricity production. Natural gas is used for a variety of purposes including electricity production, chemical production, industrial process heat and space heating. Liquid fuels used for transportation and coal consumption for electricity production are each responsible for 30% of US CO_2 emissions. Current levels of atmospheric CO_2 are about 350 ppm. International organizations are working toward agreements aimed at stabilizing that level in the future at no more than 550 ppm.

Table 1 Current Energy Generation Scenario (Bose-"Energy, Global Warming, and Impact of Power Electronics")

Resource	United States, % of Total	Global, % of Total
Oil	41	38
Natural Gas	23	31
Coal	23	28
Nuclear	8	6
Renewables	5	7

Table 2 shows the relatively limited future availability of the world's fossil and nuclear fuel resources. Coal will remain the largest energy resource for the next 200 years. Note also that, according to this and other reasonable future usage scenarios, the world will have exhausted its fossil and nuclear fuel resources within a period of only 600 years (approximately 1600-2200).

Table 2. Fuel Resource Depletion Estimates (Bose-"Energy, Global Warming, And Impact Of Power Electronics")

Resource	Estimated Period of Peak Production	Estimated Peak Annual Production Rate (in 10^{18} J)	Estimated Years of Supply Remaining
Coal	**2050-2070**	**1450**	**200**
Gas	**2040-2060**	**220**	**150**
Oil	**2010-2030**	**140**	**100**
Uranium	**2000-2020**	**100**	**50**

Figure 1 illustrates the predicted distribution of fuel usage for electricity production in the US through 2030. The use of renewables is expected to grow at an accelerated rate, coal use will continue to increase, while nuclear and natural gas use will grow slowly between 2007 and 2030. Cleaner, more efficient ways to utilize the coal resource should be developed by the US particularly since the United States holds about 25% of the world's coal reserves.

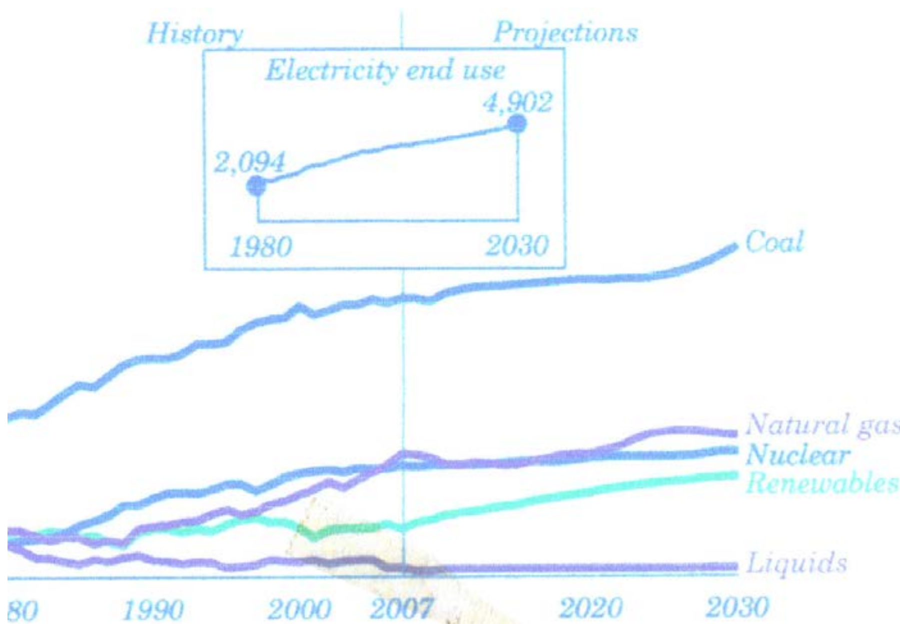

Figure 1. US Electricity Generation by Fuel. 1980-2030, billion kilowatt hours (EIA Annual Energy Outlook, Early Release Overview, January 2009)

3. Development of Technology for the Electrochemical Conversion of Carbon to Electricity

The use of a solid particulate carbon fuel in a Direct Carbon Fuel Cell (DCFC) maximizes the conversion efficiency of carbon chemical energy into electricity. Using a fuel cell means avoiding the Carnot cycle efficiency limitation of heat engines.

The maximum efficiency of a fuel cell η_t is limited by the ratio of Gibbs energy change, ΔG, of the fuel cell reaction to its enthalpy change, ΔH, or, in other terms, the loss of entropy, ΔS, in fuel oxidation reaction at operating temperature *T*:

$$\eta_t = \frac{\Delta G}{\Delta H} = 1 - \frac{T\Delta S}{\Delta H} \quad [1]$$

Using carbon fuel in solid form avoids the efficiency limitation due to entropy loss since the net DCFC reaction (C + O_2 = CO_2) has nearly zero entropy change. Therefore, the DCFC has a maximum conversion efficiency of 100% (heat of combustion of carbon to electrical energy), which is significantly higher than the maximum carbon-based efficiency of other fuel cells, which consume gaseous or liquid fuels.

This was already realized in 1894 by Ostwald, the father of energy conversion thermodynamics, who wrote: "Once we have an electrochemical cell that yields electricity directly from coal and air, then we stand on the threshold of an industrial revolution that will dwarf the one that followed upon the invention of the steam engine."

Despite these advantages, early attempts to obtain "electricity directly from coal" remained futile because of the complex kinetics of fuel oxidation, electrode inactivation by impurities from coal, and inability to supply the solid fuel efficiently to the electrodes. These barriers are easily visualized from the detailed stoichiometry of the electrode reactions:
The reaction on the anode side of the DCFC, in its simplest stoichiometry, is complete oxidation of carbon by oxygen ions with the release of four electrons and CO_2:

$$C + 2O^{2-} \rightarrow CO_2 + 4e^- \quad [2]$$

The reaction on the cathode side of the DCFC, in its simplest stoichiometry, is the reduction of oxygen molecules (from air) into oxygen ions with consumption of electrons:

$$O_2 + 4e^- \rightarrow 2O^{2-} \quad [3]$$

The problem of supplying solid fuel efficiently can be minimized in DCFC systems with a "liquid anode", that is, a suspension of solid fuel (carbon) particles in molten alkali metal carbonates. Suitable solid fuels in large quantities, at relatively low cost can be derived from widely available sources such as coal, biomass, and petroleum coke – or by pyrolysis or other thermal treatment of agricultural, food processing, and municipal solid waste. Contaminants in carbon fuel derived from these sources affect the fuel cell kinetics to different degree, but various ways to minimize or avoid this poisoning effect have been demonstrated. The development issues that remain to be resolved are considered in Section 4.

If these technological barriers are overcome, a system efficiency level can be achieved that would be between 1.5 and 2 times that of combustion or gasification systems in large central station power plants. In contrast to these central systems, the production cost of electricity by a DCFC based decentralized system is projected to be lower than grid-delivered electricity because it avoids transmission and distribution charges. However, the low cost fuel, high conversion efficiency, and modest capital cost of the DCFC plant itself (targeted at $2000-2500/kW) are the principal reasons why a lower COE is expected to be achievable.

The DCFC is expected to be deployed in modular, distributed, stand-alone 0.3-10 MW power plants that can be located to supply electricity consumed on site at locations where particulate solids are collected as the result of manufacturing or waste disposal operations carried out for other purposes. CO_2 emissions from electricity production plants are related to the efficiency of the conversion process and the composition of the fuel. Biomass-based fuels are typically considered to be CO_2-neutral when used as fuel for electricity production.

Summarizing, the major attribute of DCFC technology that is relevant to low-cost electrical energy production as well as reduction of CO_2 emissions, and economic and environmental benefits, is that it has the potential to convert chemical energy of particulate carbon directly to DC electricity at 60–65% overall system efficiency in a DCFC power plant. The product gas is >95% pure CO_2 and the efficiency penalty for CO_2 capture and compression is less than 5%. Therefore, DCFC technology has the potential to maintain coal as an environmentally acceptable fuel for power generation.

The potential advantages of DCFC for production of a substantial fraction of US electricity from coal and carbon-rich solid wastes include:

- Reduction of imports of liquid fuels from foreign sources by providing lower cost electricity to power the conversion of the US auto fleet from gasoline to electric power by using the US coal reserves in an environmentally acceptable way
- Reduction of the amount of CO_2 produced per kWh of electricity produced
- Reduction of the cost of capturing CO_2 from DCFC based power plant effluent streams
- Establishing a new technological lead for the US in advanced energy technologies.

4. DCFC Technology Development Issues

The major development risk is whether the technical barriers can be overcome at an acceptable cost with a system approach that can produce competitive electricity compared with alternative approaches. There are several design alternatives, which will affect efficiency, lifetime, and reliability at various levels of the fuel cell assembly. These alternative designs involve (1) the geometric configuration of the liquid anode, O^{2-} conducting layer, and oxygen electrode (cathode) which together form a cell or module, that is, a repeating element with a certain nominal voltage ; (2) the number of cells combined in parallel, or number of cathodes configured with one liquid anode, to form a module of certain current capacity; (3) the number of modules which are combined electrically in series to form a stack, or fuel cell assembly, of desired power capability. All of these design alternatives must be considered when addressing DCFC system scale-up. For example, to minimize ohmic losses in interconnects the fuel cell assembly should have, for a given power output, the highest possible DC voltage and lowest possible DC current.

The major challenges involve (1) the performance of the solid cell elements (cathode layer plus O^{2-} conducting layer) in terms of power density and, to a lesser extent, efficiency, (2) the rate of performance decline and ultimate (estimated maximum) life of the solid cell elements; (3) the cost of the fuel cell assembly . Electrical efficiency related design parameters are ohmic (IR) losses in interconnect and liquid anode composition and convection intensity. Among the system efficiency related parameters is the power required for auxiliary equipment. Lifetime and reliability related design parameters, which indirectly impact cost, include: overall simplicity of the design (especially minimizing the number of moving parts), sensitivity to failure of individual stacks, and ability to replace stacks or other components.

Specific targets for fundamental research in DCFC systems that could achieve transformational results by the end of the TIP funded research efforts include:

- Identification of more active cathode, solid-electrolyte, and anode current collector materials that can operate at 600-700C, that is, 100-200C lower than currently tested systems. Lower temperature operation reduces and may eliminate the rate of corrosion of all solid component materials of the DCFC. Naturally this also requires long-term testing of DCFC modules that incorporate new materials.
- Increased electronic conductivity of the liquid anode. Like any fuel cell anode, the DCFC anode must function as a mixed conductor. Molten salts are not electronically conductive. Electronic conductivity in the DCFC “liquid anode” occurs by “percolation” from one carbon particle to another, and therefore may be enhanced by inert but conducting additives.
- Improved current collector effectiveness (for example, by configuration) and life.
- Testing and mitigation of the effect of contaminants such as sulfur (and COS), chlorine, and mineral matter on anode processes and anode current collection.

5. DCFC Development Pathways

DCFC emissions of contaminants in the fuel can be controlled easily in accordance with regulations, and contaminated salt from the anode chamber can either be purified or disposed of in accordance with RCRA requirements for contaminants originally present in the fuel. However, as mentioned, a major threat to maintaining DCFC performance, in terms of power output over time, is poisoning (deactivation) of cathode or solid-electrolyte, and/or corrosion of the current collector by contaminants in the fuel.

Standard prerequisites for acceptance by the electric utility industry for large central station power generating applications are: (1) successful operation in terms of sustained performance; (2) maintenance of full-scale component modules for long operating periods, with Forced and Planned Outage Rates that are comparable to today’s coal-fired units. Considering the effect of contaminants, both of these prerequisites suggest that the most prudent development pathway for DCFC technology development is to focus on small scale applications (50 kW to few MW). Notably, other types of stationary fuel cells (MCFC, PEM, PAFC, and SOFC) have found market niches in this range. These systems are fueled either with hydrogen or natural gas that is converted to hydrogen upstream of the fuel cell anode. In the DCFC case analogous fuels would be particulate carbons derived from biofuel, food, and wood processing wastes because these materials are low in sulfur, chlorine, and mineral matter. Development of larger scale systems capable of handling Municipal Solid Waste residues would be the next step. Coal would follow after smaller systems achieved successful long-term operation on less difficult fuels.

6. DCFC Fuel Supplies

The US currently utilizes over 1 billion tons of coal per year to generate slightly over 2000 billion kWh of electricity. It is highly desirable to start, now, reorienting this industry to technologies with high conversion efficiency that reduce both fuel consumption and CO_2

emissions. The DCFC would provide an optimal basis for this, with US coal supplies more than adequate for at least the next two centuries.

Both solid-biomass fuels (crop residues, farm waste, food processing waste, MSW, sludge waste, and wood/wood waste) and industrial waste (petroleum coke, textile mill residue) are potential major sources of solid carbon fuel. For example, according to the United Nations Statistics Division, Energy Statistics Database, in 2005 U.S. industries' solid waste products amounted to 105,100 TJ (equivalent to 1×10^{14} Btu). Assuming that this amount of energy could be converted in DCFC systems to electricity at a system efficiency of 65%, 2,200 MW would be produced hourly throughout the entire year. Another major source of waste energy is MSW. The U.S. EPA has estimated that in the United States the annual production rate of MSW is 4.5 lb per person. It has been estimated (Wolk, R., Lux, S., Gelber, M., and Holcomb, F.: *Direct Carbon Fuel Cells: Converting Waste to Electricity*. ERDC/CERL TR-07-32. 2007) that the amount of recoverable carbon in MSW is about 20%. Utilization of all of this resource in the United States for electricity production via DCFC would produce about 30,000 MW of electricity hourly throughout the entire year.

A 2006 report to the California Energy Commission (*Assessment of Biomass Resources in California* (PIER Collaborative Report, Contract 500-06-016; December 2006) estimated the potential for electricity generation from biomass-derived fuels in California at 3,900 MW. This estimate was based on 2005 production quantities of waste biomass and 30% conversion efficiency in conventional combustion systems. Using DCFC in place of combustion and achieving a 65% system conversion efficiency would increase the potential to about 8,400 MW of continuous power production throughout the year. Approximately 3,600 ton/yr of biomass-derived waste (assuming a heating value of 13.3 million Btu/ton) is required to produce 1 MW continuously throughout the year at 65% conversion efficiency. The amount of waste generated is proportional to the population. On a national scale, based on the ratio of population in California relative to the national population, the U.S. market for these systems should be in the range of 5-10 times larger than the California market.

Thirty-one states in the US have established Renewable Portfolio Standards (RPS); those RPS stipulate differing amounts of, and implementation dates for, the renewable-based generation required. Assuming that DCFC capture a 1% share of the RPS market in 2015, 5% in 2020, and 10% in 2025, a market would exist of 196 MW by 2015, 1,869 MW by 2020, and 6,018 MW by 2020.

8. Transformational Aspects of the DCFC Technology

According to the World Coal Institute, in 2006, 41% of the electricity generated in the world was produced from coal. This compares sharply with the 2.3% produced from all types of renewable energy. Coal is likely to remain as the primary fuel worldwide for electricity production for decades to come, since it relatively low in cost and is the world's largest fossil fuel resource with a projected remaining supply of 200 years. Unfortunately, from an environmental standpoint almost all of this electricity was produced by burning coal and emitting the resulting CO_2-rich flue gas into the atmosphere. What is needed to be able to use this resource in the future in an environmentally responsible manner is a markedly improved technology.

Table 3, which includes information from EPRI Report 1016170, April 2008, compares the fuel-to-bus-bar electricity generation efficiency, estimated CO_2 emissions, capital cost, and Cost of Electricity (COE), with and without CO_2 capture for two state of the art technologies- Supercritical Pulverized Coal (SCPC) for combustion power plants, and Integrated Coal Gasification (IGCC), as well as two fuel cell based developmental technologies - Integrated Gasification Solid Oxide Fuel Cell (IGSOFC) and DCFC. It should be noted that the estimate of capital investment required and resulting Cost of Electricity for each of those technologies were calculated on a consistent basis

Table 3. Estimated Efficiency, CO_2 Emissions, Capital Costs of and Cost of Electricity from Coal Fired Plants (EPRI Report 1016170, April 2008

Technologies	**Efficiency, Fuel to busbar electricity, % HHV**		**CO_2 Emissions, mt/MWH**		**Estimated Capital Costs, $/kWh**		**COE, $/MWH**	
	No CO_2 Capture	**CO_2 Capture**	**No CO_2 Capture**	**CO_2 Capture (% capture)**	**No CO_2 Capture**	**CO_2 Capture**	**No CO_2 Capture**	**CO_2 Capture**
SCPC	38	27	0.86	0.12 (90)	2290	3820	55	99
IGCC	38	37	0.83	0.11 (90)	2720	3780	66	97
IGSOFC	50	43	0.65	0.08 (90)	2200	3046	52	77
SRI DCFC	65	60	0.49	0.00 (100)	2323	2750	52	67
CellTech DCFC	65	60	0.52	0.00 (100)	2494	2963	56	71
Contained Energy * DCFC	65	63	0.49	2.26 (49)	2136	2368	63	73

*Only this technology case used de-ashed coal; all other cases use raw coal

Almost all of the world's coal plants in service today utilize pulverized coal combustion to produce electricity by means of a high pressure steam cycle. These plants are typically in the 300-800 MW capacity range. The CO_2 leaves the power plant as an atmospheric pressure flue gas with a CO_2 concentration of 12-15%. CO_2 capture from this gas is energy intensive and expensive as indicated in Table 3 by the large loss in efficiency. It is not economically feasible to capture more than 90% of the CO_2 in that flue gas. An attractive technology option for new coal-fueled power plants is Integrated Gasification Combined Cycle (IGCC). In these plants, coal is gasified, the product gas cleaned, and then burned in a combined cycle (gas turbine plus steam turbine) unit. If CO_2 capture is required, the plant can be configured to remove CO_2 and other contaminants at high pressure. That clean gas, consisting primarily of hydrogen is then burned in a combined cycle to produce electricity. This approach results in a much lower loss of efficiency and lower incremental cost compared to pulverized coal power plants. At this time, the largest plants of this type that have been built have a capacity of 250-300 MW. A number of projects for 600 MW power plants are being planned

One of the major current DOE programs to improve the efficiency of coal powered power plants is based on the use of Solid Oxide Fuels Cells (SOFC). In this approach, the same product gas that can be produced in an IGCC plant is converted to electricity in SOFC. This is a technology that exists on a laboratory scale and will require future scale-up and demonstration.

According to the information summarized in Table 3, only DCFC technology has the potential to achieve 60% efficiency with 100% CO_2 capture at a COE and with an estimated capital investment requirement that is lower than any conventional or developmental alternatives.

9. Likely Proposers for DCFC R&D

Organizations that have been active in various areas of DCFC technology development and are likely proposers to perform R&D work in this area include: 4D Power LLC, Contained Energy, CellTech Power, SRI International, Direct Carbon Technology, Hawaii Energy Research Institute, Rocketdyne, Nextech, Ceramatec, and Satcon.

10. Need for Federal R&D Support

Consideration of these challenges that have been identified in previous and on-going R&D efforts related to DCFC leads to the conclusion that long-term, well funded development programs will be required to achieve commercialization. As a result, private sector investors have been deterred by the anticipated long-time horizon required to achieve a reasonable return on their R&D investments. Federal government funding is needed to accelerate the further development of this promising electrochemical technology through the modular component demonstration phase. The private sector will then have an acceptable basis to support further scale-up to economically viable commercially-scale systems.

DCFC has the potential to be an important future commercial power production technology that can reduce the consumption rate of fossil fuels, utilize renewable fuels at high efficiency, and reduce CO_2 emissions (or CO_2 sequestration costs by reducing the amount of CO_2 produced.)

Delaying its further development means that its future commercial availability will be delayed for a comparable period. Such delay in deployment of truly clean and highly efficient technologies for distributed/localized power generation from coal may have implications for social instability and economic uncertainty. The closest competitor, natural gas power plants, is being shut down as a result of volatile prices. The only realistic substitute power source for natural gas is coal. In the absence of clean coal technologies, conventional or “advanced” coal combustion or gasification technologies have to be deployed with negative impact on the environment. Clearly, environmental concerns will limit deployment of combustion technologies, which, in turn will limit electricity production growth and slow down such vital undertakings as migration of transportation to “all electric” option and favor continued dependence on foreign oil. The situation with natural gas power plants may be considered as a lesson: the key factor for predictable electricity production is guaranteed fuel supply. There is no other guaranteed fuel source for large scale power generation, which is remotely comparable with coal. After all, the question is quite simple: Nature blessed the US with coal – will we continue to burn it, or can we do something better?