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Fuel Processing Technology 71 (2001) 7–38

FUEL  
PROCESSING  
TECHNOLOGY

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# Review of advances in combustion technology and biomass cofiring

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## Abstract

Advances in combustion technology will be adopted only when they reduce cost and can be implemented with acceptable technical risk. Apart from technical risk, future decisions on new power plants will be principally influenced by trends in fuel cost, the efficiency and capital cost of new generating technologies, and environmental and regulatory policies including possible carbon taxes. The choice of fuel and generating technology for new power plants is influenced by an increasingly complex combination of interrelated factors: (1) current and future governmental policies on restructuring and deregulation of utilities, and environmental regulations that in the future could include taxes on carbon emissions; (2) macroeconomic factors such as proximity to load centers, electrical transmission lines, plant capital investment, delivered fuel cost, and fuel price stability; and (3) the state of development of new generating and environmental control technologies and the associated benefits and risks involved in their deployment, which are strongly related to fuel properties. This paper describes three advanced high-efficiency power systems for which the EERC has performed supporting research and development: (1) a coal-fired supercritical steam boiler with advanced emission controls; (2) an indirectly fired combined cycle using compressed air as the working fluid in a gas turbine (GT), fired either on coal alone or on coal and natural gas; and (3) two versions of a hybrid gasifier-pressurized fluidized-bed combustor (PFBC) system. © 2001 Elsevier Science B.V. All rights reserved.

*Keywords:* Combustion technology; Biomass cofiring; Fuel

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## **1. Introduction**

The Energy and Environmental Research Center (EERC) is a nongovernmental research center within the University of North Dakota established to advance the science and technology of energy conversion and utilization and environmental protection in partnership with private industry and government agencies. We believe that strong research partnerships committed to scientific and engineering excellence throughout the development cycle are the key to technological progress, and we welcome collaboration on jointly sponsored research with organizations throughout the world. Prior to privatization in 1983, the EERC was the US Department of Energy's (DOE's) lead laboratory for US low-rank coals, which in the United States include large reserves of lignite and subbituminous coal in western and Gulf Coast states comprising over half of the total US coal tonnage resource. Our research has since expanded to include projects worldwide that encompass all fossil fuel resources (coal, oil, and gas), renewable energy (biomass and wind), advanced power systems, ambient air quality and source emission controls, basinwide water management and water quality, and environmental remediation (nuclear and nonnuclear). We believe that attention to fuel quality is the key to successful application of new and advanced energy conversion technologies, which builds on our historic focus on highly diverse low-rank coals. We have recently expanded our research to include an emphasis on biomass fuels and particularly on cofiring biomass with coal as a practical, near-term approach to reducing net emissions of carbon dioxide, which are a concern because of forecasts of global warming. In this review paper, we will (1) critique some of the coal properties that have a major influence on their utilization, (2) describe three advanced combustion systems within the DOE program on which the EERC is performing research and development related to coal properties, (3) examine issues involved in cofiring biomass, and (4) review the emerging issue of toxic trace element emissions from coal combustion.

## **2. Fuel properties and generating costs**

The choice of fuel and generating technology for new power plants is influenced by an increasingly complex combination of interrelated factors: (1) current and future governmental policies on restructuring and deregulation of utilities, and environmental regulations that in the future could include taxes on carbon emissions; (2) macroeconomic factors such as proximity to load centers, electrical transmission lines, plant capital investment, delivered fuel cost, and fuel price stability; and (3) the state of development of new generating and environmental control technologies and the associated benefits and risks involved in their deployment, which are strongly related to fuel properties. This paper focuses on the third category, but first considers some general effects of fuel properties on representative generating efficiencies and costs and relative carbon emissions.

Table 1 compares average properties and cost estimates for competing fossil and biomass fuels, including natural gas, oil, US coals of all ranks, Turkish and Bulgarian

lignites, and three types of biomass. Prices given for US coals are the 1997 average delivered prices for coal mined in the respective states as published by the DOE Energy Information Agency (DOE EIA [52]), while those for other fuels are approximations based on general knowledge of resource recovery and market conditions. Prices for natural gas in the United States have recently become extremely volatile in the range of \$5 to \$10 and even higher. If gas prices stabilize at about \$5/mscf, gas will continue to be a competitive choice for new power generation in gas turbine (GT) combined-cycle systems that offer efficiencies over 50% (higher heating value [HHV]) at a capital cost of approximately \$600/kW and deliver a cost of electricity (COE) of about \$32/MW h, or 3.2 cents/kWh. The fuel cost component of electrical generation using coal is considerably lower than that for natural gas, even for conventional pulverized fuel (pf)-fired plants operating at efficiencies between 26% and 34% (HHV) or between 33% and 35% (lower heating value [LHV]), depending on moisture and ash content. US practice customarily defines efficiency on the basis of the HHV, which is the value obtained when the water in combustion products is condensed, whereas European practice uses the LHV, which recognizes that water in practice is not condensed. The difference is approximately 10% of the efficiency for hydrogen-rich natural gas, i.e., 50% (HHV) versus 55% (LHV), only 2.4% of efficiency for a low-moisture bituminous coal and up to about 25% of efficiency for a high-moisture, high-ash lignite. This difference must be taken into consideration when comparing published efficiencies and evaluating the effect of coal properties on efficiency.

The COE calculations in Table 1 indicate that some low-cost coal even when utilized at the efficiency of conventional pulverized coal (pc)-fired plants, is today fully competitive with 85/msol natural gas burned in a GT combined-cycle. This comparison is based on a simplified economic analysis that assumes (1) a cost for a steam plant that is twice that of a GT combined cycle, (2) a 10-year payback (not using discounted cash flow analysis), (3) a similar plant capacity factor of 80%, and (4) fixed cost apart from capital changes and operating and maintenance costs apart from fuel of \$9, \$5, and \$3/MW h for coal, oil, and gas, respectively. The COEs for biomass costing \$40/short ton are higher than those for either coal or gas. In today's deregulated markets, natural gas plants have the advantage of low capital cost where merchant generating plants need to attract capital by providing a rapid payback. However, prices for natural gas have been extremely volatile, and are projected to rise more rapidly, than those for coal. The large reserves and relative price stability of coal give it a competitive advantage over time in the absence of policy disincentives. In addition, new technologies of the types reviewed in this paper are projected to reduce the capital costs of advanced coal-fired plants by up to 20% and to increase efficiency levels to 45–50%, which would result in the substantial reductions in COE that are estimated in Table 1 and discussed in the conclusion of this paper.

In the future, coal could be severely disadvantaged in relation to natural gas and renewable energy resources if policy instruments such as carbon taxes are implemented to discourage carbon emissions from power plants. Relative carbon emissions are presented in Table 1 and Fig. 1, comparing natural gas to other fuels both at equivalent generating efficiency and at the characteristic LHV efficiencies of current technology. This comparison indicates that the carbon emission from high-moisture, high-ash lignites

Table 1  
Comparison of cost and emission factors for selected fuels<sup>a</sup>

	Natural gas 96v% CH <sub>4</sub>	US Pennsylvania bituminous	US Illinois bituminous	US Wyoming subbituminous	US North Dakota lignite
<i>Fuel cost factors</i>					
Fuel units	mscf	short ton	short ton	short ton	short ton
Estimated fuel cost at source, \$/unit <sup>b</sup>	\$5.00	\$33.28	\$30.41	\$14.16	\$10.21
Energy cost, \$/MM Btu HHV	\$3.02	\$1.30	\$1.23	\$0.74	\$0.76
Characteristic generating efficiencies, % of HHV <sup>c</sup>	50.0%	33.4%	33.2%	32.5%	30.9%
Characteristic generating efficiencies, % of LHV <sup>c</sup>	55.0%	34.6%	34.5%	34.5%	34.2%
Fuel cost per MW h at characteristic efficiencies HHV	\$34	\$13.26	\$12.69	\$7.79	\$8.38
COE at characteristic efficiency, \$/MW h <sup>d</sup>	\$46	\$39	\$39	\$34	\$34
Target efficiency for a supercritical boiler, % HHV		42.8%	42.7%	41.7%	39.8%
COE for supercritical boiler at \$1300/kW capital cost <sup>e</sup>		\$38	\$37	\$32	\$34
Target efficiency for hybrid gasifier–PFBC, % HHV		45%	45%	45%	45%
COE for advanced power system at \$900/kW capital cost		\$32	\$31	\$26	\$28
<i>Impact of an energy tax of \$50 per ton of carbon</i>					
Increase in fuel cost, \$/unit	\$0.79	\$35.82	\$32.72	\$27.45	\$20.02
Increase in energy cost, \$/MM Btu in fuel HHV	\$0.79	\$1.40	\$1.33	\$1.44	\$1.49
Increase in \$/MW h at characteristic efficiency	\$5.40	\$14.28	\$13.65	\$15.10	\$16.42
Cost of fuel plus tax, \$/MW h	440	\$28	\$26	\$23	\$25
COE with carbon tax at characteristic efficiency, \$/MW h <sup>d</sup>	451	\$54	\$52	\$49	\$51
COE with tax for advanced systems at 45% HHV and \$900/kW		\$42	\$41	\$38	\$39
<i>Averaged proximate analysis and heating value, wt.% as received basis</i>					
Moisture	0.0%	3.5%	3.70%	18.9%	37.6%
Volatile matter		34.7%	37.40%	34.8%	26.3%
Fixed carbon		50.9%	44.20%	39.3%	29.7%
Ash	0.0%	10.9%	14.70%	7.0%	6.4%
HHV, Btu/lb	23 106	12 840	12 320	9550	6730
LHV, Btu/lb	20 814	12 271	11 739	8847	5867
LHV, kcal/kg	11 563	6817	6522	4915	3260

*Averaged ultimate analysis, wt.% moisture and ash-free basis<sup>f</sup>*

Carbon	72.4%	83.7%	80.20%	74.1%	71.5%
Hydrogen	24.1%	5.6%	5.70%	5.3%	4.9%
Oxygen	0.0%	6.7%	7.00%	18.0%	21.3%
Sulfur	0.0%	2.5%	5.40%	1.0%	1.1%
Nitrogen	3.5%	1.5%	1.70%	1.6%	1.2%

*Averaged ash analysis, wt.% of oxides in ash on a CO<sub>3</sub> and SO<sub>3</sub>-free basis corrected to total 100%*

SiO <sub>2</sub>		49%	49%	48%	27%
Al <sub>2</sub> O <sub>3</sub>		30%	20%	19%	14%
Fe <sub>2</sub> O <sub>3</sub>		15%	20%	12%	12%
CaO		3.5%	6.8%	16.0%	31.0%
MgO		0.6%	0.9%	4.1%	8.8%
Na <sub>2</sub> O		0.6%	0.5%	0.7%	6.6%
K <sub>2</sub> O		1.3%	2.0%	1.0%	0.6%
P <sub>2</sub> O <sub>5</sub>					

*Calculated adiabatic flame temperature (assumes 100% burnout of fuel at 21°C), 20% excess air, and an air preheat temperature of 300°C*

	1975	2057	2041	1951	1769
<i>Sulfur dioxide emissions (assuming 100% emission of fuel sulfur)</i>					
lb SO <sub>2</sub> /MMBtu HHV	0.01	3.33	7.15	1.55	1.83
g SO <sub>2</sub> /Mcal LHV ar	0.018	6.28	13.51	3.02	3.78
mg SO <sub>2</sub> /dscm	12.2	4143	9146	1939	2265
Ratio of SO <sub>2</sub> to EEC standard of 400 mg/dscm	0.03	10.36	22.87	4.85	5.66
<i>Carbon dioxide emissions</i>					
g CO <sub>2</sub> /Mcal LHV ar	229	385	368	410	450
Ratio of CO <sub>2</sub> emissions/CO <sub>2</sub> emission for natural gas					
Assuming equivalent generating efficiency	1.00	1.68	1.60	1.79	1.96
Assuming characteristic LHV efficiencies <sup>c</sup>	1.00	2.67	2.56	2.85	3.16

*(continued on next page)*

Table 1 (continued)

	Australia morwell brown coal	Bulgaria maritsa east lignite	West Turkey lignite	East Turkey lignite	Air dried willow wood	Straw	Wood waste
<i>Fuel cost factors</i>							
Fuel units					short ton dry	short ton dry	short ton dry
Estimated fuel cost at source, \$/unit <sup>b</sup>					\$40.00	\$40.00	\$14.00
Energy cost, \$/MM Btu HHV					\$2.52	\$3.09	\$0.97
Characteristic generating efficiencies, % of HHV <sup>c</sup>					31.9%	31.0%	26.9%
Characteristic generating efficiencies, % of LHV <sup>c</sup>					34.5%	34.4%	33.5%
Fuel cost per MW h at characteristic efficiencies HHV					\$26.91	\$33.97	\$12.38
COE at characteristic efficiency, \$/MW h <sup>d</sup>					\$53	\$60	\$39
Target efficiency for a supercritical boiler, % HHV					40.9%	39.8%	34.7%
COE for supercritical boiler at \$1300/kW capital cost <sup>e</sup>					\$49	\$54	\$37
Target efficiency for hybrid gasifier–PFBC, % HHV					45%	45%	45%
COE for advanced power system at \$900/kW capital cost					\$41	\$45	\$29
<i>Averaged proximate analysis and heating value, wt.% as received basis</i>							
Moisture					6.4%	9.6%	47.60%
Volatile matter					75.2%	74.4%	42.39%
Fixed carbon					14.5%	8.4%	0.68%
Ash					3.9%	7.6%	9.33%
HHV, Btu/lb					7441	5858	3766
LHV, Btu/lb					6785	5149	2682
LHV, kcal/kg					3769	2860	1490
<i>Averaged ultimate analysis, wt.% moisture and ash-free basis<sup>f</sup></i>							
Carbon					48.5%	41.45%	51.09%
Hydrogen					6.2%	6.40%	6.12%
Oxygen					44.1%	51.08%	41.75%
Sulfur					0.0%	0.25%	0.03%
Nitrogen					1.2%	0.82%	1.00%

Averaged ash analysis, wt.% of oxides in ash on a  $CO_3$ - and  $SO_3$ -free basis corrected to total 100%<sup>f</sup>

SiO <sub>2</sub>	0.9%	21.1%	5.7%
Al <sub>2</sub> O <sub>3</sub>	0.1%	1.6%	2.3%
Fe <sub>2</sub> O <sub>3</sub>	1.0%	1.2%	2.0%
CaO	42.4%	26.3%	63.3%
MgO	6.5%	5.5%	7.3%
Na <sub>2</sub> O	0.3%	1.1%	4.6%
K <sub>2</sub> O	33.1%	31.8%	10.5%
P <sub>2</sub> O <sub>5</sub>	15.7%	11.4%	4.4%

Calculated adiabatic flame temperature (assumes 100% burnout of fuel at 21°C), 20% excess air, and an air preheat temperature of 300°C

1913	1805	1419
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Sulfur dioxide emissions (assuming 100% emission of fuel sulfur)

lb SO <sub>2</sub> /MMBtu HHV	0.52	12.63	7.09	10.51	0.07	0.70	0.07
g SO <sub>2</sub> /Mcal LHV ar	1.23	35.70	14.66	26.13	0.14	1.44	0.12
mg SO <sub>2</sub> /dscm	648	14907	8762	12553	94	949	90
Ratio of SO <sub>2</sub> to EEC standard of 400 mg/dscm	1.62	37.27	21.90	31.38	0.24	2.37	0.22

Carbon dioxide emissions

g CO <sub>2</sub> /Mcal LHV ar	526	606	438	541	423	440	542
Assuming equivalent generating efficiency	2.29	2.64	1.91	2.36	1.85	1.92	2.36
Assuming characteristic LHV efficiencies <sup>c</sup>	3.76	4.43	3.08	3.94	2.94	3.07	3.88

<sup>a</sup>Fuel analyses are taken from Selle et al. [41] for US coals and file analysis for wood.

<sup>b</sup>Fuel prices for US coals are the average delivered price of coal mined in the state published by US DOE [54]. Estimates of other fuel costs are approximations based on general knowledge of resource recovery and market conditions and are used only to illustrate differences in generating costs and carbon tax impacts.

<sup>c</sup>The 50% HHV generating efficiency for natural gas represents a current state of the art gas turbine-combined cycle. Efficiencies for coal and wood are for a steam turbine generator and were calculated using an EERC boiler model for steam conditions of 540°C and 180 bar with one cycle of reheat to 540°C.

<sup>d</sup>Cost of electricity is based on capital costs of \$600/kw for a combined cycle and \$1200/kw for a steam plant, with an 80% plant factor and a 10-year straight payback. Fixed charges other than capital plus maintenance and operations apart from fuel are estimated at \$9/MW h for coal, \$5/MW h for oil, and \$3/MW h for gas.

<sup>e</sup>COE based on a capital cost of \$1300/kw and the target efficiency for a supercritical boiler, all other assumptions are the same as for footnote “d”.

<sup>f</sup>The ash analysis for willow wood is taken from Skrifvars et al. [42].

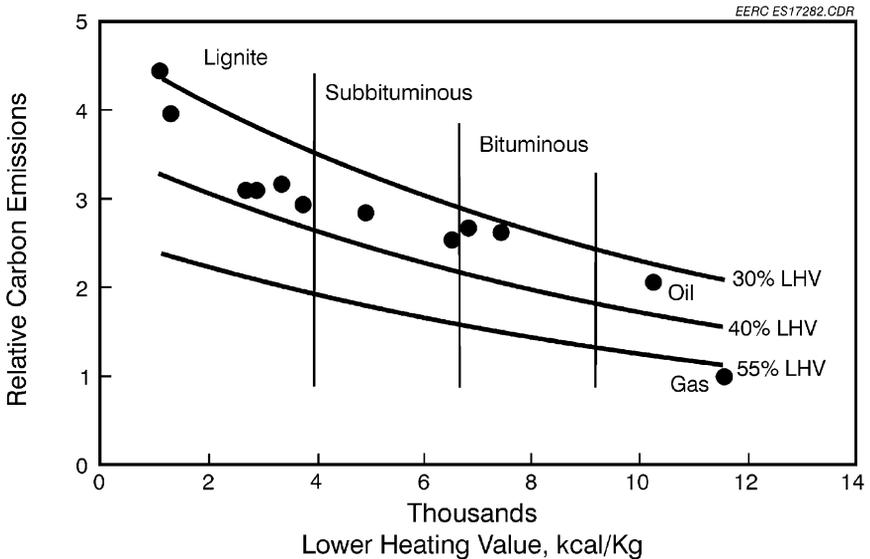


Fig. 1. Effect of fuel type and generating efficiency on relative carbon emissions.

fired in a conventional pc-fired boiler would be approximately fourfold higher than the emission from a natural gas-fired combined-cycle plant, based on fuel stoichiometry and LHV efficiencies. This difference could have immense economic significance if a carbon tax were imposed. A 1997 DOE [54] study prepared by the DOE National Laboratories [33] estimated that a carbon tax of \$50/short ton of fuel carbon content would be required to bring US CO<sub>2</sub> emissions back to their 1990 level, as required by the Kyoto protocol, which the United States has yet to ratify. Much higher costs have been estimated by some other studies. If such a tax were to be necessary, it would add between \$14 and \$20/MW h to the cost of coal-based electricity (a 30% to 40% increase), compared to only a \$5/MW h increase for natural gas fired in a combined cycle (about 11%). Coal would then only marginally be competitive with natural gas within the current price structure, and biomass would become less expensive than coal if it were not subject to the tax. This scenario may overstate the influence of policy in the near future, but it illustrates the importance of developing highly efficient advanced power systems for coal and investigating the practicality of cofiring biomass to reduce net carbon emissions. The other technical solution for dealing with carbon emissions from continued and expanded utilization of coal, which appears essential to economic growth in many parts of the world, would be the development of methods for sequestering the CO<sub>2</sub> produced. These methods will not be discussed here. The following sections of this paper will describe three advanced high-efficiency power systems for which the EERC has performed supporting research and development: (1) a coal-fired supercritical steam boiler with advanced emission controls; (2) an indirectly fired combined cycle using compressed air as the working fluid in a GT, fired either on coal alone or on coal and natural gas; and (3) two versions of a hybrid gasifier-pres-

surized fluidized-bed combustor (PFBC) system. All of these advanced systems depend on higher temperatures to achieve improved thermodynamic efficiencies, which raises questions regarding high-temperature corrosion and deposition by coal ash and slag. The cofiring of some types of biomass may also encounter deposition problems owing to the occurrence of unique forms of silica and the much higher concentrations of potassium and phosphorus found in their ash.

### 3. The supercritical low-emission boiler system

The low-emission boiler system (LEBS) developed under the sponsorship of DOE [53] integrates a state-of-the-art supercritical boiler with advanced stack gas-cleaning technologies to achieve a net generating efficiency exceeding 42% HHV (45% LHV) with emissions no greater than 0.1 lb NO<sub>x</sub>/MMBtu (124 mg/scm), 0.1 lb SO<sub>2</sub>/MMBtu (124 mg/scm), and 0.01 lb particulate/MMBtu (12 mg/scm). These emission levels are 1/3 to 1/10 of the current US New Source Performance Standards, and approximately 1/4 the level of the European Community (EC) standard for large coal-fired plants. After several years of design and subscale testing by three boiler manufacturers, the D.B. Riley design was selected in 1997 to move toward construction of a “proof-of-concept” power plant. Construction of this plant in Illinois is awaiting final funding arrangements by sponsors. Alternative designs were developed by Babcock and Wilcox and ABB-Combustion Engineering. The D.B. Riley design incorporates a low-NO<sub>x</sub> slag-tap boiler with low-temperature heat recovery; a regenerable, moving-bed copper oxide adsorber for NO<sub>x</sub> and SO<sub>2</sub> control; and a pulse-jet fabric filter for particulate capture (DOE [55]). This paper does not discuss the stack gas-cleaning methods used by D.B. Riley, but supercritical boiler technology will be reviewed.

The current state of the art for coal-fired supercritical steam plants is represented by the 400-MW Nordjyllandsvaerket plant in Denmark, which was placed in service in 1998 [24]. Steam conditions for this plant are 285 bar (4130 psia) and 580°C (1076°F), with double reheat to 580°C. Net design efficiency when burning a high-grade bituminous coal was reported to be 47% LHV (44% HHV) for a 105°C (221°F) boiler exit temperature and a cooling water temperature of 10°C (50°F) using seawater. Boiler efficiency under these conditions would be approximately 95% LHV (92% HHV). EPRI has performed studies for similar steam conditions of 310 bar (4500 psia) and 593°C (1100°F). Future state-of-the-art steam temperatures are projected to increase to 620°C (1148°F) by 2005 and to 650°C (1202°F) by 2020 [13], with each step up adding ca. one percentage point in efficiency to reach 49% LHV (46% HHV) for the boiler exit and cooling water conditions of the Nordjyllandsvaerket plant.

Apart from steam conditions, the efficiency of a supercritical steam plant is most sensitive to boiler exit temperature, cooling water temperature, and the moisture and ash levels in the coal. Increasing the stack temperature from 105°C (221°F) to a more usual lower limit of 125°C (257°F) would reduce the efficiency by about a 0.5 percentage point. Raising the cooling water temperature from a very low 10°C (50°F) available to a coastal power station using seawater up to 21°C (70°F) for tower cooling would reduce the efficiency by an additional 1.4%, bringing the design efficiency of the Nordjyl-

landsvaerket plant down to about 45% LHV (42% HHV) for an inland site in a moderate temperature zone, which is the same as the year 2000 efficiency goal set by DOE for its LEBS program. Because of stack losses, the boiler efficiency of a plant operating on lignite will drop several percentage points and plant efficiency will be significantly reduced. For the analysis of North Dakota lignite used in Table 1, with 38% moisture and 6.4% ash on an as-received basis, boiler efficiency is reduced to 93% LHV (84% HHV) and net generating efficiency is 40% HHV, assuming a 125°C boiler exit temperature and 21°C cooling water. For West Turkish lignite with 25% moisture and 26% ash, boiler efficiency is calculated to be 92% LHV (85% HHV), and efficiency is 44% LHV (41% HHV).

The main problem limiting the application of supercritical steam-generating plants over a range of coal qualities is high-temperature fireside corrosion by alkali-iron-tri-sulfates that form a molten layer on tube surfaces at temperatures between approximately 620°C and 700°C [28]. This problem would be expected to be a particular concern for firing some lignites, which contain high concentrations of both sodium and sulfur and for cofiring biomass with a high concentration of potassium along with a high-sulfur coal (refer to Table 1). Considerable progress has been made through international cooperation in developing new materials with sufficient creep strength and resistance to steam side oxidation for steam temperatures in the range of 600°C and higher. However, materials need to have adequate resistance to fireside corrosion at temperatures approximately 35°C higher than the design steam temperature. Information on high-temperature fireside corrosion is limited, and materials tests now being conducted are mainly on natural gas and low-sulfur coals [28]. Extensive materials research that is under way cannot be reviewed within the scope of this paper. However, in general the rate of fireside corrosion for ferritic materials increases linearly with increasing temperature, whereas the rate for austenitic materials peaks at about 663°C (1225°F) in the range where the corrosive alkali sulfates are most stable [28]. The use of optimized austenitic stainless steels is suggested for controlling fireside corrosion at steam temperatures up to about 620°C [24], and more highly alloyed steels under development may allow for higher steam temperatures of 630°C (1166°F) [13].

The EERC has performed laboratory corrosion tests on alloys for supercritical boilers under test conditions representing the cofiring of a high-alkali, high-sulfur coal with a high-potassium biomass [22]. The test alloys were selected based on conversations with materials engineers from Elsam, the Danish utility that constructed the Nordjyl-landsvaerket plant, and were all high-Ni–Cr alloys considered suitable for steam temperatures up to 630°C (1166°F). The laboratory procedure involved covering alloy samples with a mixture of Illinois bituminous coal ash (see Table 1) and potassium sulfate and heating to 625°C in a synthetic combustion gas containing 1000 ppmv SO<sub>2</sub> for 100 h or longer. Water vapor alone and in combination with HCl were added in later tests performed over a period of 1000 h. The principal finding from these tests was that alloys, which formed an iron oxide layer experienced considerably more corrosion than those that formed a chromium oxide layer. The alloy exhibiting the least corrosion had the highest chromium content (HR3C), and the alloy showing the most corrosion had the lowest chromium and highest nickel content (Incoloy 800HT). The effect of adding water vapor on corrosion rates was minor. However, the addition of HCl to the gas

stream increased the depth of corrosion significantly. Alloys containing less than 20% chromium were most affected, with corrosion depths up to eight times greater when the HCl was added to the gas stream [22].

#### 4. High-performance power systems

After the supercritical low-emission boiler, the technology that next most closely resembles a conventional pc-fired boiler is the DOE high-performance power system (HiPPS) being developed by a team headed by the United Technologies Research Center (UTRC). This technology integrates a combustion GT and heat recovery steam generator (HRSG) with an advanced coal-fired boiler in a combined-cycle power system that can be fueled either on coal alone or on a combination of coal and natural gas. The unique feature of the HiPPS cycle, as shown in Fig. 2, is that air from the GT compressor is heated in a coal-fired high-temperature advanced furnace (HITAF). The heated compressed air from the HITAF can either be used directly to operate the GT or the air can be additionally heated by burning natural gas to increase cycle efficiency. Variations of the basic cycle based on different steam conditions and GT temperatures have been proposed for new or retrofit applications. The basic design offers estimated gross efficiencies (HHV) between approximately 42% for a GT high-pressure temperature (GTHPT) of 982°C (1800°F) burning coal alone up to 47% for a GTHPT of 1371°C (2500°F), with natural gas providing 35% of the heat input [48]. Still higher gross efficiencies between 54% and 56% (HHV) are being projected for more advanced cycles [37,38], using either (1) a humid air turbine, which saturates the air from an intercooled compressor (after it passes through an aftercooler) to increase mass flow through the

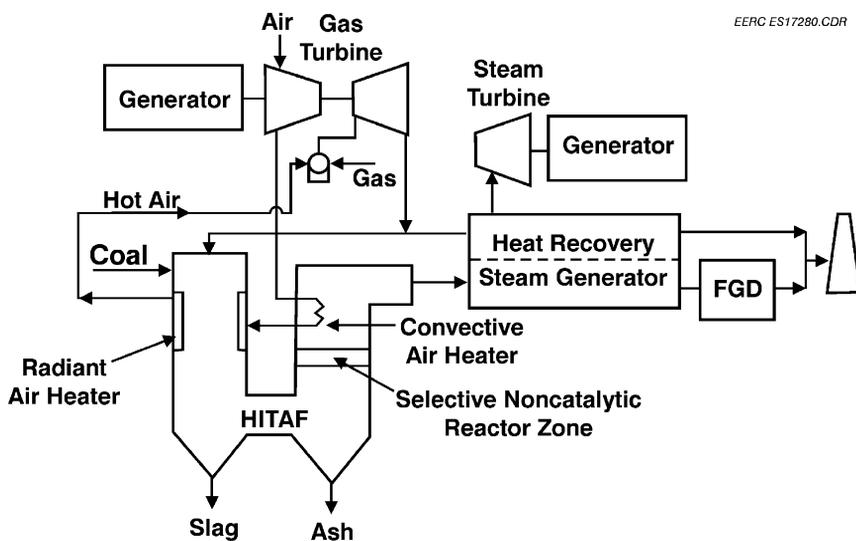


Fig. 2. Schematic of the UTRC HiPPS cycle.

turbine or (2) a developmental solid oxide fuel cell in series with an advanced high-temperature (1491°C [2716°F]) GT. UTRC has calculated the cost of a green field plant using the basic HiPPS technology to be \$1038/kW, which it estimates to be 8% below the cost of a new pc-fired plant and 30% higher than the cost of a natural gas-fired combined-cycle plant [39].

The EERC in partnership with UTRC and PSI Technology Company has designed, constructed, and operated a 3.0-million Btu/h ( $3.2 \times 10^6$  kJ/h) slagging furnace system (SFS) for the purpose of validating design concepts, materials, and heat-transfer coefficients for the HITAF. The principal test components of the SFS system as shown in Fig. 3 are (1) a high-temperature slagging furnace designed to operate at fireside temperatures between 1482°C and 1593°C (2700°F to 2900°F), (2) a slag screen consisting of water-cooled high-alumina ceramic-coated stainless steel tubes arranged in a staggered pattern to capture ash particles by impingement before they enter the convective air heater (CAH), (3) a dilution and quench zone where the gas temperature is reduced to about 1010°C (1850°F) by recirculating stack gas, and (4) the high-alloy (e.g., incoloy) CAH, which heats compressed air to temperatures approaching 705°C (1300°F). The heart of the HITAF is the radiant air heater (RAH, Fig. 4) that is positioned in the slagging furnace to heat the compressed air leaving the CAH from about 705°C (1300°F) to the design temperature of 982°C (1800°F). The RAH consists of a series of vertical heat exchange tubes made of a specially formulated nickel-based oxide dispersion-strengthened alloy (MA 754) that is protected from slag corrosion by refractory plates made from fusion cast alumina. The test program on the EERC's SFS has validated the basic HITAF design concept including the heat-transfer regime, and has exceeded the design target by heating compressed air to 1093°C (2000°F), which is believed to be a world record for a coal-fired system.

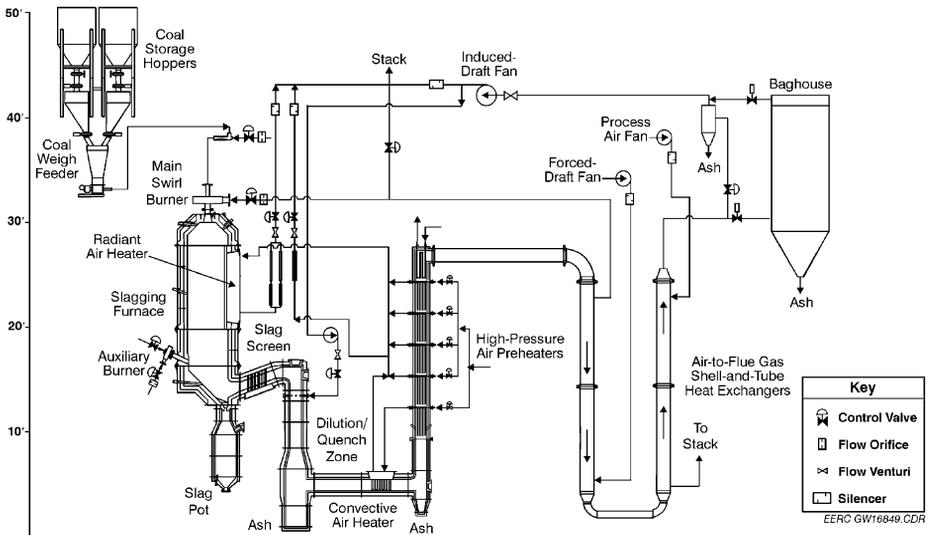


Fig. 3. EERC SFS used in the development of the UTRC HITAF.

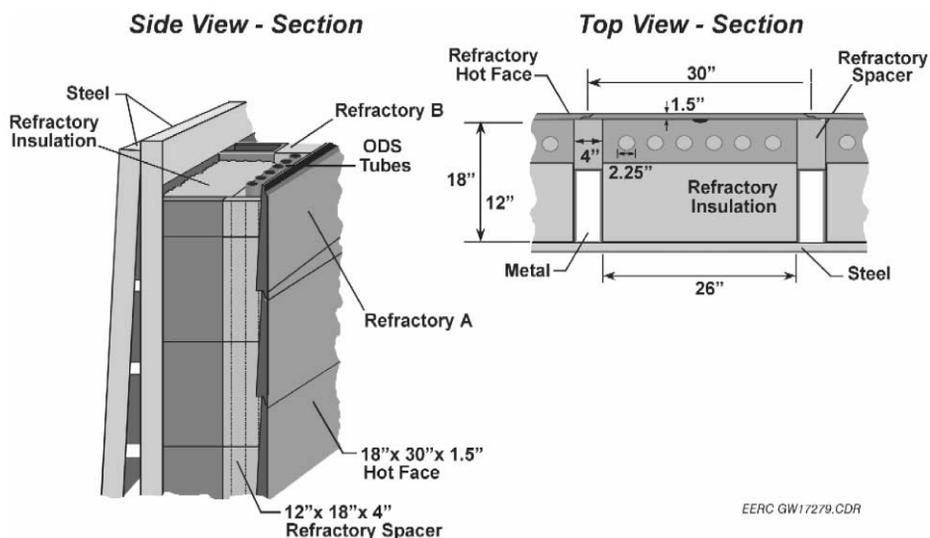


Fig. 4. RAH for the UTRC HITAF.

The technical issues that will affect the application of the HiPPS technology are concerned with the effect of coal quality on efficiency, operability, and the resistance of materials to slag corrosion. The EERC slagging furnace has been successfully operated on six US coals of different rank: two bituminous coals from Kentucky and one from Illinois, a Wyoming subbituminous coal, and two North Dakota lignites. Cycle analysis performed by UTRC indicates that the HiPPS incurs a 2% efficiency penalty (HHV) when firing a 35% moisture lignite in place of bituminous coal [10], which is essentially the same as the penalty that we at the EERC have calculated for a supercritical boiler. Peak furnace temperatures experienced when burning lignite in the SFS were lower, owing to the higher moisture content of the fuel. The most significant problems with operability concern plugging of the slag screen and difficulty in removing slag, accentuated in the smaller dimensions of the SFS compared to a large commercial unit, which were successfully controlled by adjusting temperature and employing additives to modify slag properties. Ash deposits that formed on the alloy tubes in the CAH were limited to the leading and trailing edges and did not impede gas flow. Deposits affected heat transfer as in a conventional pc-fired boiler, with sootblowers expected to successfully mitigate ash deposition and maintain desired heat transfer in a commercial unit. In general, the HITAF design provides the same fuel flexibility as a conventional pc-fired boiler, except that the exposure of the alloy heat exchangers and ceramic materials of construction to much higher temperatures accentuates the importance of slag corrosivity.

An extensive evaluation of ceramic materials for use in the HITAF furnished significant results [10,48–51]. The fusion cast alumina refractory (Monofrax M) plates used to protect the alloy tubes in the RAH of the SFS experienced a corrosion rate higher than would be acceptable for commercial use, especially along slag flow paths. During 1005 h, total exposure to all of the various coal ash slags, similar discoloration

due to slag penetration was observed, limited to about 6 mm. However, problems with castable refractories were found to be more coal-specific. Laboratory corrosion tests performed at the EERC using a specially designed dynamic slag application furnace (DSAF) were used in cooperation with the Plibrico Company to develop the high-alumina castable refractory lining of the slagging furnace that was used successfully for Illinois and Kentucky bituminous coal slag. However, the new castable experienced rapid corrosion by lignite-type slag, which was confirmed in the SFS tests on lignite. Aged areas of castable refractory in the SFS that had been previously coated with slag from previous test runs using bituminous coal were not degraded, indicating that a high-fusion-point coating may be helpful in protecting the refractory from corrosion by corrosive lignitic slag. However, coatings that were tested were not shown to either reduce the corrosion of castable refractory in the DSAF or improve the heat-transfer characteristics of the ceramic tiles used in the RAH. A sintered chrome-alumina refractory brick manufactured by Kyocera was shown in tests on the DSAF to be substantially more resistant to slag corrosion than the fusion cast alumina (Monofrax M), and operational tests on a Kyocera RAH tile are ongoing in the SFS. Other DSAF tests showed that prefiring an experimental 98% alumina castable refractory to 1625°C (2957°F) reduced the corrosion rate by 75% compared to material prefired to only 1500°C (2732°F). Extensive fundamental research was performed at the EERC in conjunction with the HITAF project to evaluate slag corrosion of additional materials, including sintered  $\beta$ -SiC, silicon nitride, and molybdenum disilicide, and to understand how changes in slag viscosity due to crystallization affect mass transport mechanisms controlling corrosion rates close to the temperature of critical viscosity (the temperature at which flow changes from Newtonian to Bingham plastic viscosity behavior) [20–23].

## 5. Hybrid gasifier-pressurized circulating fluidized-bed systems

The hybrid combined cycle shown in the simplified schematic in Fig. 5 integrates the partial gasification of coal with pressurized circulating fluidized-bed (PCFB) combustion in a gas-turbine combined cycle that is fueled entirely on coal. This concept was the basis for a Clean Coal Demonstration project proposed to DOE in 1992 by the EERC, Babcock and Wilcox, and Minnesota Power and Light to use North Dakota lignite [60], and the Low-Rank Coal Cooperative Research Center in Australia is working on a similar technology for its brown coal. Continuing developments sponsored by DOE are reviewed below.

In all of the various designs, coal is partially gasified with air (and possibly some steam) at typically 870° to 930°C (1600° to 1700°F) and 10 to 23 bar to produce a low-Btu gas, and the residual char is burned in a PCFB using a high level of excess air. The low-Btu gas is then combined with the hot gas from the PCFB in a topping combustor to raise the gas temperature to 1260°C (2300°F) or higher for expansion through the GT. Net generating efficiencies of 46% to 49% HHV are reported for the technologies supported by DOE, depending on the GT inlet temperature and steam cycle conditions [11,36,44]. The efficiency of this type of system increases as a larger fraction of the total power is generated in the GT, all other factors being equal. We at the EERC



(1400°F) ahead of the candle filters to minimize filter plugging [57], and the fuel gas also passes through a packed bed of emathelite pellets to remove alkali vapors before being burned in the topping combustor. Steam is generated in both the PCFB heat exchanger and a HRSG to drive the steam turbine. The topping combustor uses swirl burners that aerodynamically create fuel-rich and fuel-lean zones to minimize  $\text{NO}_x$  formation. Dolomite is injected into the gasifier to catalyze tar cracking and to capture sulfur as calcium sulfide, which is later transformed to calcium sulfate in the PCFB operating at high excess air. Based on pilot plant tests, 95% sulfur capture can be achieved at a 1.75-calcium-to-sulfur molar feed ratio [36]. The Foster Wheeler system is designed to burn a substantial proportion of the coal carbon in the PCFB (possibly ca. 50% for bituminous coal) and to generate only sufficient low-Btu gas to raise the gas temperature to the level desired for the GT. The pilot-scale system has been tested on both bituminous and subbituminous coals, and a feasibility study has been performed for lignite [35]. It is generally known that lignites, because of their high reactivity, can be gasified at temperatures at least 100°C lower than bituminous coals, but under very mild conditions and low gasifier conversions, the gasification products from lignite contain high levels of  $\text{CO}_2$  and a low calorific value. Therefore, it would be expected that a higher fraction of the lignite would be gasified to meet the temperature requirements of the turbine, and that additional lignite would be fed to the PFBC to generate sufficient steam for the Foster Wheeler design.

The same type of cycle as shown in Fig. 5 has been proposed for a system using the M.W. Kellogg transport reactor, which uses a very compact, fast fluidized bed with a high heat release rate capable of operating in both combustion and gasification modes [58]. This reactor is expected to lower the cost of a topping-PCFB combined-cycle system, and it may be particularly well suited for highly reactive coals such as lignite. Preliminary estimates of capital cost range from under \$900/kW [45,60] to \$1072 [11] for a technology projected to be available in about 2007.

Since 1995, the EERC has tested the Kellogg transport reactor on all ranks of coal including lignite in a 140-kg coal/h transport reactor development unit (TRDU) operating in both gasification and combustion modes [46]. A schematic of the EERC facility is shown in Fig. 6. Coal is fed along with dolomite through a lockhopper and screw feeder into the lower mixing section of the reactor riser, where it reacts with air (and superheated steam during gasification) within a very short mean solids residence time ranging from approximate 20 to 40 s, depending on the diameter of the mixing zone. The solids are separated at the top of the reactor in both a disengager and cyclone and drop back to the bottom of the reactor for recycle or are discharged. Carbon conversion is essentially completed in one cycle during combustion, but continues over a number of cycles for gasification. The hot gas is directed to the hot-gas filter vessel, which can test up to 19 candle filters up to 1.5-m long.

Limited testing of the EERC's transport reactor in the combustion mode has yielded excellent results. Carbon conversion was 99% at 873°C (1603°F) and 8.6 bar for a nonreactive petroleum coke [46]. Sulfur capture was 92.3% at a Ca/S ratio of 1.0 [46].  $\text{NO}_x$  levels averaged 100 ppm (about 210 mg/scm), or about 1/3 of the EC emission standard for large coal-fired plants. The  $\text{N}_2\text{O}$  emission was 20 ppm, which represents a fourfold reduction compared to an atmospheric fluidized-bed combustor.

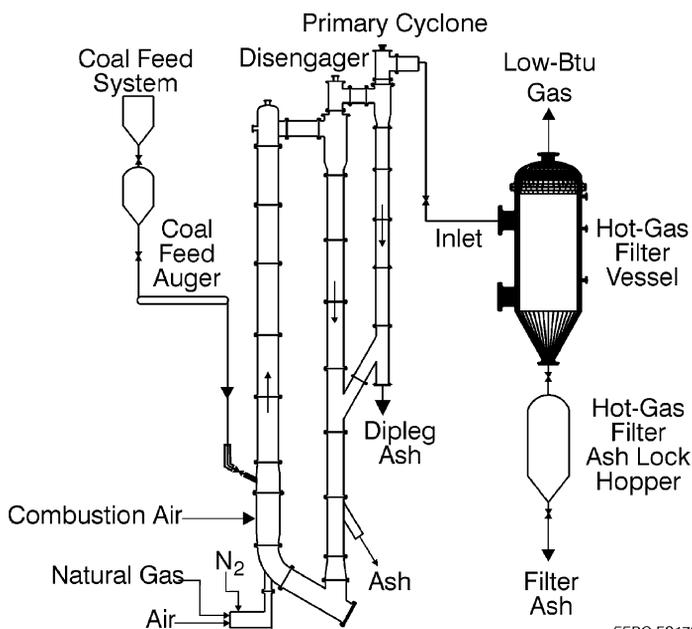


Fig. 6. Schematic of the EERC TRDU.

Over 1000 h of gasification operation have been performed on the EERC's TRDU testing seven different feedstocks, including petroleum coke, Illinois and Utah bituminous coal, Wyoming subbituminous coal, and most recently three different lignites [45]. An extensive database has been collected on the gasification performance of these fuels and on the effectiveness of design changes intended to increase carbon conversion and calorific value of the gas. The gasifier carbon conversions on coal calculated from the analysis of the solids residue were in the range of 70–90% and were higher for low-rank coal. Optimum gas heating values corrected to the conditions of a commercial reactor were in the range of 113–130 Btu/scf and did not appear to be affected by coal rank [45]. Operation was generally characterized to be more trouble-free for subbituminous coal and lignite, although some ash deposition was experienced for all coals. The most troublesome deposits formed in the mixing zone where air first contacts the bed carbon and where localized hot spots can occur. Increasing the solids circulation rate improved gas solid mixing and reduced deposition. The chemical composition of the mixing-zone deposits reflected the composition of the coal ash, with the iron-rich deposits formed from Illinois coal being harder and more difficult to control than the softer and more porous calcium aluminosilicate deposits formed from the Wyoming subbituminous coal and Utah bituminous coal (see Table 1 for a typical analysis of Wyoming and Illinois coals). Detailed examination of deposits from the riser and disengager sections from testing of Wyoming subbituminous coal, using computer-controlled scanning electron microscopy (CCSEM), indicated that the bonding materials consisted of a calcium- and

magnesium-rich coating on the surfaces of the bed material, similar in composition to the smaller particles that were retained in deposits on candle filters [7].

Since 1997, a larger 650-kg/h Kellogg transport reactor has been operated in combustion mode for over 2700 h at pressures up to 15 bar (220 psia) at the DOE Power Systems Development Facility at Wilsonville, AL [56]. Gasification tests are currently under way. Wilsonville combustion tests have produced results very similar to those obtained at the EERC, with 99.9% carbon conversion on bituminous coal,  $\text{NO}_x$  emissions of about 120 ppm, and nearly complete sulfur capture on dolomite [56].

## 6. Hot-gas cleaning

The principal technical obstacle to deployment of the topping-PCFB combined cycle and other related PFBC and integrated gasification combined-cycle technologies is the lack of confidence in the reliability of hot-gas cleaning for removing particulates from the gas stream to protect turbine blades. After decades of development, opinions still differ as to the state of development of ceramic filters. Materials and engineering problems appear to have been largely resolved, but problems relating to the filter bridging and blinding behavior of the ash remain, with numerous reports of cake buildup on filter elements that has been difficult, if not impossible, to remove. Bridging involves the formation of ash deposits that bridge between filter elements and cause them to break or the vessel outlet to plug, because of the stickiness of the ash. Bridging is the more serious problem in combustors because it limits the temperature at which the vessel can operate. Surface blinding is the more serious problem at the lower temperatures at which gasification filters often operate. In 1994, the EERC began a 3-year research effort on the problem of filter bridging under the sponsorship of DOE, EPRI, and a consortium of eight commercial groups from Europe and the United States concerned with commercializing hot-gas-cleaning technology [14]. This research was directed toward combustion applications and included the characterization of filter ash from most of the pressurized combustors operating in Europe and the United States. At the same time, tests were carried out on the EERC's TRDU to evaluate hot-gas filter performance under gasification conditions [46].

The EERC's work on filter blinding [14] involved a fundamental approach to predicting the sticking and sintering of ash particles resulting from liquid and solid bonds formed by condensation of alkali salts and gas–liquid–solid reactions involved in the formation of sulfates and silicates. The work relied on the EERC's extensive background in characterizing coal mineralogy and ash formation using advanced CC-SEM and thermochemical equilibrium modeling and included measurements of ash partitioning related to variations in ash size and composition with position in a filter vessel and in the filter cake. Characterization was performed on filter cake ash from the AEP Tidd project and other large experimental PFBCs, and these results were augmented by performing tests on the EERC's bench-scale pressurized fluidized-bed reactor equipped with a small hot-gas filter vessel to measure fragmentation and coalescence of ash components and the effectiveness of alkali getters. The primary variables tested were

cake temperature, cake porosity, particle-size distribution, in situ chemical reaction, the presence of liquid phases on particle surfaces, and water vapor effects. Initial sulfation of the ash was observed to occur within 5 min of depositing on the ash cake, which implies a need for frequent on-line cleaning by backpulsing. Since ash bridges must support themselves against gravity, tensile strength in relation to the density of the cake is an important parameter. In order to make the extensive data on many different aspects of this problem available to system engineers, results were reduced to a series of algorithms which have been assembled into a computer code, the Filter Bridging Index Code. This proprietary code uses input data from the analyses of the coal and sorbent along with system conditions to first calculate the particle-size and composition distributions of the ash entering the filter vessel and then calculate the relative stickiness of the ash resulting from liquid sulfates and low-viscosity silicates. The EERC is endeavoring to extend this work and would welcome inquiries from interested parties.

Hot-gas filter tests under gasification conditions were carried out in the EERC's TRDU at temperatures between 540°C to 650°C (1000°F to 1200°F) and 8 bar and at a filter face velocity of 2.5 ft/min [46]. Inlet particulate loadings ranged from 3500 to 34,000 ppm, and the high-carbon-content ash (ca. 50% C) had a low bulk density of 20 lb/ft<sup>3</sup> (0.32 g/cm<sup>3</sup>). The low-density filter cake lacked cohesiveness, and significant reentrainment was experienced after backpulsing. No candle failures occurred in 650 h of testing, with only a slight loss in candle permeability after nearly 6000 backpulses. However, the baseline "clean" pressure drop increased in some tests, from 20 to over 80 in. (50 to 200 cm) of water. This increase in the baseline indicated that a thin but low-permeability cake remained on the filter surface after backpulsing. Injection of filter aid additives reduced but did not eliminate the rise in baseline pressure drop. Fundamental research of the type described above for combustion systems is needed to understand the behavior of ash particles from gasification in filter cakes and the forces retaining a residual cake on the filter after backpulsing.

## 7. Impacts of cofiring biomass with fossil fuels

Coal burning electric utilities in the United States are becoming interested in biomass cofiring as a practical, relatively low-cost option for reducing greenhouse gas emissions, taking advantage of the fact that biomass fuels close the carbon cycle by converting the solar energy stored via photosynthesis into thermal and electrical energy. However, widespread adoption of cofiring may still be some distance in the future, depending on government policy. The DOE reference case estimate of biomass use for power generation given by the EIA is 1.5% of coal-based electricity by the year 2020 [52]. EPRI has estimated that 2.29% of coal generation could be displaced at a net cost of \$22.62 per metric ton of carbon above the cost of coal, using biomass priced under \$0.96/MM Btu [19]. The eventual potential for biomass cofiring where the fuel is available may be considerably larger, since the thermal input from biomass to individual boilers can be in the range of 5–15% or possibly higher, depending on boiler design and biomass-feeding method. The economics of cofiring are also benefited by the value of

tradable emissions credits under the US caps on SO<sub>2</sub> and NO<sub>x</sub> emissions. However, with the exception of wood waste used close to its source of supply, most sources of forest or agricultural biomass can be expected to cost \$2 to \$3/MMBtu, which is significantly higher than the cost of coal (see Table 1). Therefore, widespread biomass cofiring by utilities will likely not occur in the United States without policy incentives or carbon taxes. There is, however, a growing interest among utilities in marketing “green power” as customers become more aware of the forecasts of global warming and are empowered to choose electricity from renewable resources at a premium price in a deregulated retail power market. There is also a very high level of research interest in test firing biomass, including the programs of DOE, EPRI, the Tennessee Valley Authority, and other utilities in the United States [19,47]. This paper will review some of the significant findings from US and European research and present information on research activities that have been initiated at the EERC.

Past biomass—only boilers have generally been small and inefficient, and the delivered cost of biomass fuels, other than wood waste, has been high because of their dispersed supply, low energy density, and expensive handling. Cofiring provides a more practical and cost-effective means of utilizing biomass by taking advantage of the relatively high efficiency of large utility boilers without incurring a large capital investment. In the future, the advanced power systems described in this paper can utilize biomass cofiring at higher efficiencies. Biomass cofiring also faces some risks and limitations, which in individual cases may include deficiencies in facilities for storing, feeding, and pulverizing the fuel; adverse fireside impacts on flame stability, ash fouling or slagging, corrosion, and carbon burnout; and disruptions in the operation of emission control devices for SO<sub>2</sub>, NO<sub>x</sub>, and particulate. Cofiring different types of biomass containing a variety of mineral constituents not found in coal along with a diversity of coal types creates unique combustion problems that must be identified and corrected to avoid discrediting biomass cofiring as a practical utility option.

Many different types of biomass are available for cofiring, including (1) wood wastes from pallets, telephone poles, sawdust, and manufacturing scraps; (2) agricultural remnants from peach pits, rice hulls, wheat straw, alfalfa, rape, barley, soybeans, sunflowers, bagasse, and other grains; (3) residues from logging, orchards, and forest management; (4) fast-growing energy crops such as hybrid poplar, willow, black locust, eucalyptus trees, and switchgrass; and (5) municipal wastes including plastic, paper, and cardboard. A supply infrastructure for gathering, transporting, storing, reclaiming, and delivering biomass is a critical requirement for large-tonnage use of biomass, as is quality control. An EERC literature survey of biomass analyses identified a very wide range of properties: moisture contents from < 10% to over 50%, ash contents from < 1% to 20%, nitrogen contents from 0.1% to over 2% (4% for one plywood waste), and sulfur contents from near zero to ca. 0.3%. Chlorine content varies from near zero to 0.6%. Biomass has a high ratio of volatile matter to fixed carbon (typically 60–80% volatile matter to 10–20% fixed carbon dry), and between 30% and 50% oxygen content on a moisture- and ash-free (maf) basis. The maf hydrogen content is only slightly higher than for coal. The combustible in biomass can be viewed as the end of a progression from bituminous coal, to lignite, to biomass. Emissions of carbon dioxide from biomass in relation to heat release are in the same range as for lignite (see Table

1), and biomass use must be viewed in relation to the closed photosynthesis cycle in order to obtain credit for reducing greenhouse gas emissions (a credit that is sometime challenged). The three biomass analyses included in Table 1 also illustrate a large variability in the major oxide constituents in the ash analysis: silica ranging from 1% to 21%, calcium oxide from 26% to 63%, potassium oxide from 10% to 33%, and phosphorus pentoxide from 4% to 16%. These ranges, which do not encompass the full variability in biomass ash analysis, differ substantially from the ranges in the analysis of coal ash, particularly with respect to the much higher concentrations of potassium and phosphorus in the biomass. High concentrations of silica, potassium, and chlorine occurring in some herbaceous biomass represent a special concern for deposition and corrosion in boilers.

Key cost and operating factors for cofiring have been presented in recent studies [12,19]. An important limit is the fraction of biomass that can be fed through pulverizers in a pc-fired plant, which is less than 4% by mass, or about 2% by heat input for a plant using bituminous coal. Higher percentages of cofiring require a separate feed system for feeding biomass directly into the boiler through separate injection ports. Cyclone boilers are more adaptable since the coal is crushed rather than pulverized, although maintaining the proper cyclone temperature and slag viscosity will impose limits. The capital costs for retrofitting biomass cofiring used in the EPRI study [19] were \$40 to \$50/kW for blending through the crusher (for cyclones) or the pulverizer (for pc-firing), and \$175 to \$230/kW for feeding higher percentages of biomass through a separate feed system. Other capital cost estimates fall in a range of \$50–\$400/kW, where higher costs represent difficult handling situations [12]. Cofiring rates up to 15% of heat input for medium pc-fired boilers are reported in cost studies, but the maximum cofiring rates in DOE-EPRI-sponsored boiler tests have been 10% of heat input for both cyclones and pc boilers (wall- and tangentially fired) [19]. The overall economics of cofiring can be shown to be potentially profitable in special circumstances where low-cost, locally available biomass displaces high-cost, high-sulfur coal. In the most favorable cases, the leverage of increased dispatching in a deregulated power market (permitted by lower fuel cost) plus emission credits of \$1.71/MW h for SO<sub>2</sub> and \$3.23/MW h for NO<sub>x</sub> are shown to generate rates of return over 100% on the capital investment for retrofitting [12]. Possible future credits from a large (e.g., \$50/ton) tax on carbon emissions would tip the scale heavily in favor of cofiring economics by adding \$15 to \$20/MW h to the cost of using coal in power generation (Table 1).

Combustion properties are substantially different for various types of biomass and very different compared to coal. Among different coals, lignite and brown coal most closely resemble biomass (particularly wood). As with certain woody lignites, more energy is required for size reduction of biomass compared to bituminous coal. The high volatile matter content of the biomass burns off quickly in a boiler, and the time required for complete combustion is short compared to that for a coal particle of similar size. Past concerns over flame stability when burning highly reactive lignite in pc-fired boilers could possibly apply to biomass at high replacement percentages (this is not known to be a concern at the ca. 10% cofiring rates currently considered). Guidelines for complete combustion derived from laboratory combustion studies indicate that biomass char burnout may become a problem for top sizes greater than 3 mm and fuel moisture

contents exceeding 40% [2]. A correlation of boiler efficiency for cofiring tests at three power plants indicated approximately 0.5% loss for 10% biomass input [47]. The effect of cofiring on  $\text{NO}_x$  has varied, with reductions between 0% and 20% [19,47].  $\text{NO}_x$  reduction can result from several factors, including reduced fuel nitrogen content, lower firing temperature, and staging of combustion due to early volatile burnout in the biomass fraction.

No serious increase in ash deposition was reported for tests on utility boilers in the United States [19,47] and in Finland [25] when cofiring sawdust and clean wood wastes. However, increased deposition has been reported for cofiring up to 20% sunflower hulls with North Dakota lignite [32] and for firing 20% straw with bituminous coal in Denmark [16,59]. Cofiring in Denmark was suspended owing in part to a combination of technical problems including ash deposition problems, production of fly ash that did not meet specification for use in concrete, and fouling of selective catalytic reduction (SCR) catalysts. A recent study on the properties of concrete containing wood–coal fly ash reported no significant detrimental effects on strength, workability, permeability, or setting time due to the presence of the wood ash [8]. The boiler ash deposits from cofiring straw were characterized by the presence of potassium aluminosilicates along with small amounts of  $\text{K}_2\text{SO}_4$  and by the substitution of iron for calcium in the deposit structure, apparently because of the formation of reduced iron species with relatively low viscosity [16]. In tests on a slipstream of flue gas from 20% straw cofiring, the SCR catalyst used for  $\text{DeNO}_x$  was found to be 35% deactivated in 2860 h in a high-dust environment and 15% in 2350 h in a low-dust environment after fabric filtration [59]. The deposit layer formed on the catalyst in the high-dust test was composed of mainly silica, alumina, and sulfur along with smaller amounts of potassium and phosphorus that originated from the straw. While lower deactivation rates were expected at full scale, the high cost of catalyst replacement makes this a serious economic issue.

High-temperature corrosion of boiler tubes by sulfates was reported when cofiring 20% straw in regions of the boiler reaching temperatures up to  $580^\circ\text{C}$  ( $1076^\circ\text{F}$ ) [59]. No corrosion was observed at 10% straw cofiring, and no corrosion initiated by chlorine was observed at either cofiring level. A mechanism proposed for chlorine-induced corrosion based on thermodynamic equilibrium predicts that chlorine in deposits should decrease sharply with increasing sulfur content, with little chlorine remaining on the heat-transfer surface in the presence of excess sulfur [2]. However, since more chlorine has been observed in deposits than predicted by equilibrium, the conversion of alkali chlorides to alkali sulfates is apparently limited by kinetic or transport effects [2].

Cofiring experience to date indicates that deposition and corrosion problems can vary widely when cofiring different forms of biomass with different coals. The EERC has observed similarly wide variations in ash fouling studies on high-alkali lignitic and subbituminous coals [3,4,21], which are helpful in understanding deposition behavior in cofiring biomass. These studies were discussed in an earlier paper [43]. Sodium enrichment in amorphous aluminosilicates, which results in low-viscosity liquid phases and sintering, is the foremost cause of severe ash fouling on superheater and reheater surfaces at temperatures above  $1038^\circ\text{C}$  ( $1900^\circ\text{F}$ ). At lower temperatures further back in the convection section of the boiler, the calcium in low-rank coals is sulfated to form a lattice that progressively strengthens deposits over a period of days, a process which is

accelerated at higher ratios of total alkali to silica,  $(\text{Na}_2\text{O} + \text{K}_2\text{O} + \text{MgO} + \text{CaO})/\text{SiO}_2$ . The small amount of phosphate in low-rank coals appears to affect ash fouling in the same manner as sulfate, suggesting that phosphate crystallization may be an important mechanism in ash deposition from biomass. Under locally substoichiometric combustion conditions, alkali-iron sulfides form stable low-melting eutectics, which may account for iron enrichment in biomass ash deposits [16].

The EERC is currently performing research on biomass cofiring in a bench-scale (40,000 Btu/h) pf conversion and environmental process simulator (CEPS) to advance the understanding of high-temperature ash behavior. Research is also focused on refinement of analytical methods for analyzing unique mineral and inorganic forms in biomass. CCSEM analysis reveals that the silica in wheat straw is in the form of thin, sheet-like phytoliths, which are a form not found in coal. The inorganic constituents in biomass are also being characterized by using a chemical fractionation method previously developed for lignite, which classifies the form and chemical association of elements based on their sequential solubility in water, ammonium acetate solution, and dilute hydrochloric acid. This fractionation distinguishes among (1) water-soluble salts; (2) ion-exchangeable cations associated with the organic structure; (3) acid-soluble oxides, hydroxides, and carbonates; and (4) acid-insoluble minerals such as quartz, clay, and pyrite. Chemical fractionation of the inorganic material in wheat straw indicated that (1) phosphorus, potassium, and 15% of calcium were soluble in water; (2) 65% of calcium was soluble in ammonium acetate solution; and (3) another 20% of the calcium was soluble in dilute HCl. The CCSEM analysis identified some calcium in the mineral form calcite. Silicon, iron, and titanium did not appreciably dissolve in any of the solutions, indicating that they were present as insoluble mineral forms. Combustion cofiring tests performed in the CEPS on 20% wheat straw by weight with a high-sulfur Illinois bituminous coal (see Table 1) resulted in ash fouling deposits that evidenced low-viscosity liquid phases enriched in potassium–iron aluminosilicates formed by the rapid interaction of soluble potassium with silica phytoliths and pyrite-derived iron from the Illinois coal. The CCSEM analysis of the fly ash collected from different temperature zones showed iron enrichment at high temperatures and potassium enrichment at lower temperatures, the latter indicating that potassium had condensed on the surface of the ash particles from the gas phase, either as sulfate or chloride. The finer fraction of the bulk fly ash,  $< 2.2 \mu\text{m}$ , was also enriched in potassium and sulfur, which is consistent with condensation of potassium compounds.

Cofiring tests were also recently completed at the EERC on a rice straw lignin that is the waste residue from the experimental Arkenol process, which uses rice straw cellulose as feedstock in an acid hydrolysis–fermentation process to produce ethanol. A sufficient yield of lignin along with some unconverted cellulose will be available from the Arkenol process to warrant collocating with a power plant to lower production costs for both. Tests were performed in both the bench-scale CEPS and in a 35 kg/h-pilot-scale pf-fired combustor with a heat release rate of 600,000 Btu/h. The tests were performed on blends of 5% and 10% dried lignin with a Montana subbituminous coal with 9% ash containing 39%  $\text{SiO}_2$ , 22%  $\text{Al}_2\text{O}_3$ , 7%  $\text{Fe}_2\text{O}_3$ , 24%  $\text{CaO}$ , and 1%  $\text{Na}_2\text{O}$  ( $\text{SO}_3$ -free basis). The lignin was received as a paste containing 79% moisture and having a pH between 10 and 12.5, resulting from treatment with sodium hydroxide in a silica

recovery step in the ethanol process. The lignin was air-dried to 19% moisture and pulverized in a hammer mill to a size ca. 60% minus 200 mesh, with a heating value of 2590 kcal/kg (5600 Btu/lb), 0.4% sulfur, and 30% ash. The ash was composed almost entirely of silica (56%) and sodium oxide (38%). CCSEM analysis identified the form of the silica as the characteristic silica phytoliths that are common in herbaceous plants.

The combustion of 5% and 10% blends of the Arkenol process-derived lignin with subbituminous coal gave excellent carbon burnout and easily maintained the desired furnace exit gas temperature of 1149°C (2100°F) just ahead of the fouling probes in the pilot-scale combustor. However, the very high sodium content of the lignin resulted in rapid deposit growth for both the 5% and 10% lignin blends, forming much larger deposits than for firing subbituminous coal alone. Characterization of the deposits showed that the sodium and silica in the lignin reacted with organically associated calcium and magnesium and the quartz and clay minerals in the subbituminous coal to form very low-viscosity sodium-rich silicate and aluminosilicate melts, sodium-calcium aluminosilicates (hauyne), and sodium-calcium sulfates. The fly ash was enriched in volatile sodium, phosphorus, and sulfur that had condensed on the surface of the fly ash particles. Deposits were enriched in silica. This lignin material could not be successfully cofired with coal in a utility boiler in the form in which it was received and prepared for these tests. Washing experiments have shown that the sodium content of the lignin can be reduced to 14% in the ash after several hours in a water bath, and further cofiring tests will be performed in the near future on the lower-sodium-content lignin after washing.

## **8. Toxic trace element emissions from coal combustion**

Coal combustion is a significant source of mercury and other trace element emissions, which are a regulatory concern in the United States under the hazardous air pollutant (HAP) provisions of the 1990 Clean Air Act Amendments (CAAA). The CAAA requires the US Environmental Protection Agency (EPA) to establish stationary source categories for 189 HAPs, including 11 potentially toxic trace elements found in coal (Sb, As, Be, Cd, Cr, Co, Pb, Mn, Hg, Ni, and Se). For those HAPs for which there are sources that emit in excess of 10 tons annually for any single pollutant or 25 tons for any combination, EPA must issue standards for maximum achievable control technology and then perform residual risk assessments to determine if further reductions are needed. In addition, EPA was specifically required to study HAP emissions from utility boilers and waste incinerators, with a focus on mercury.

In 1993, the EERC established the Center for Air Toxic Metals as an EPA-designated center of excellence for performing worldwide cooperative research with industry and government to provide a scientific basis for measuring and controlling emissions of toxic metals that impact human health and the environment. Research has focused on trace metal transformations in combustion and gasification, analytical methods development, computer modeling and database management, and control technologies. The program incorporates a strong emphasis on technology commercialization and education,

which has included the conference Air Quality: Mercury, Trace Elements, and Particulate Matter, held in December 1998 [5], and the following conference Air Quality II held in McLean, VA, September 19–21, 2000.

HAP emissions were measured at eight coal-fired power plants under a collaborative effort involving DOE, EPA, and EPRI, for which the EERC performed the data evaluation [30]. The results indicated that all of the trace elements listed in the CAAA, as well as organic pollutants, cyanide, and radionuclides, were all well below the 10-ton/year limit on individual pollutants. Particulate control combined with wet FGD showed > 99% control for As, Sb, Co, Pb, Mn, and Ni. Fabric filtration showed 95% control for Sb, Be, Cr, Co, Pb, Mn, and Ni. All sites showed > 90% control for Sb, As, Br, Cd, Co, Pb, Mn, and Ni. Emissions of mercury on a mass per unit energy basis ranged from 0.44 to 22 lb/10<sup>12</sup> Btu and annual emissions from 0.007 to 0.2 tons/year. The percentage of the mercury in the coal that was measured in the emissions varied widely from 6% to 120% and could not be explained by differences in emission control technologies. Emissions of halogens (chlorine, hydrogen chloride, and hydrogen fluoride) did exceed both the 10-ton/year individual limit and the 25-ton/year total HAP limit at five plants. Halogen emissions from boilers correlated well with the chlorine and fluorine contents of the coals burned, and they were shown to be captured by either wet- or dry-scrubbing technologies used for flue gas desulfurization.

Mercury is the toxic trace metal of greatest concern in coal combustion because of its volatility, its persistence and bioaccumulation in the environment, and its high neurodevelopmental toxicity. Coal combustion is now estimated to be the largest source of anthropogenic mercury in the United States, accounting for 46% of the total [55]. The EPA issued a mercury information collection request (ICR) requiring all US coal-burning utilities to analyze and report the mercury and chlorine contents of their coal during 1999. This information along with tests to measure emissions of elemental, oxidized, particle-bound, and total mercury at 88 randomly selected plants is being used by EPA to estimate annual mercury emissions from utility sources. A regulatory determination on the control of mercury from coal-fired utility boilers made in December 2000 established that controls will be required by 2001.

Emissions of trace elements and mercury can be predicted with some success from their occurrence in the fuel, transformations in the furnace, and ability to penetrate air pollution control devices [6,15,40]. Trace element emissions occur both in vapor form, as in the case of mercury, in submicron aerosols formed by vaporization and recondensation, and in association with larger fly ash particles. The complex mechanisms that control trace element transformations during combustion include ash formation by fragmentation, nucleation, coalescence, and shedding; gas and liquid diffusion; multi-component chemical equilibria; and homogeneous and heterogeneous reaction kinetics. Vaporization cannot be predicted from vapor pressure alone. The EERC has developed a TraceTran model based on experiments in a pressurized drop-tube furnace and a thermochemical equilibrium code to predict transformations of Hg, Se, As, Pb, Cd, Cr, and Ni under gasification conditions [6,16], and this code is being modified for combustion. Comparisons of trace element emissions predicted by the TraceTran computer code with measurements on the M.W. Kellogg transport reactor and hot-gas filter vessel operating at the EERC were in generally good agreement except for Se and

Cd. The lower-than-expected emission measurement for Se at the exit of the filter vessel at 500°C was due to capture on the calcium-rich sorbent used in the gasifier.

Understanding the transformations of mercury in combustion depends on a knowledge of its oxidation state and chemical speciation. During combustion in the flame region, mercury is completely converted to volatile  $\text{Hg}^0$  and subsequently partially oxidized to  $\text{Hg}^{+2}$  as the combustion gases are cooled during passage through the boiler [17,40]. The EERC has validated the Ontario Hydro method as the preferred method for measuring mercury species in flue gas to distinguish between elemental, gaseous oxidized forms, and particulate-bound mercury. This method was recommended by EPA for use in the mandated ICR tests performed on utility boilers [27]. Equilibrium calculations indicate the conversion to  $\text{Hg}^{+2}$  should be essentially complete upon cooling to 400°C, but speciated measurements on boiler emissions have ranged from 35% to 95%, indicating that kinetic limitations are controlling. The stable chemical species in flue gas leaving the boiler are theoretically calculated to include  $\text{HgO}$ ,  $\text{HgCl}_2$ , and  $\text{HgSO}_4$ , but nitrates and other forms may occur as intermediate products. Fly ash and gaseous species including  $\text{SO}_2$ ,  $\text{NO}$ ,  $\text{NO}_2$ ,  $\text{Cl}_2$ , and  $\text{HCl}$  in flue gas have been shown to have important effects on mercury speciation. The oxidation of elemental mercury in flue gas has been shown to be catalyzed by some fly ashes alone and by interactions between either fly ash– $\text{NO}_x$  or fly ash– $\text{NO}_x$ – $\text{SO}_2$ , but not by  $\text{NO}_x$ – $\text{SO}_2$  in the absence of fly ash [26]. The addition of  $\text{Cl}_2$  resulted in the highest level of mercury oxidation [26], whereas the addition of  $\text{HCl}$  either had no significant effect [26] or inhibited the capture of oxidized mercury on fly ash [18]. The addition of lime ( $\text{CaO}$ ) also inhibited mercury capture on fly ash [18]. While much has been learned about the chemistry of mercury in coal combustion systems, many apparently conflicting observations remain to be explained.

The control of mercury emissions from coal-fired boilers effectively depends on a high conversion to the oxidized  $\text{Hg}^{+2}$  form, which can be removed either in a wet scrubber used for flue gas desulfurization or on a sorbent injected upstream of either an electrostatic precipitator or baghouse. Tests on the injection of many types of carbon sorbents have been funded by DOE and EPRI at carbon-to-mercury ratios between about 10,000 and 100,000, which corresponds to ca. 1% to 10% of the coal ash for a 10% ash coal [58]. The required ratio will depend on the particle size, reactivity, and capacity of the sorbent, as well as the point of injection and residence time in the gas stream. Calculations on diffusion-limited adsorption indicate that a 95% reduction in an initial mercury concentration of  $10 \mu\text{g}/\text{m}^3$  can be accomplished in 2-s residence time at a carbon-to-mercury ratio of about 1000 using a sufficiently reactive carbon with a uniform particle size of  $2 \mu\text{m}$  [9]. However, the calculated carbon-to-mercury ratio increases sharply for larger particle sizes (ca. 20,000 at  $10 \mu\text{m}$ ) and for particle-size distributions with an appreciable coarse fraction, indicating that mercury adsorption can be diffusion limited at low injection rates for a sorbent having an insufficiently fine size distribution. The EERC has performed bench-scale tests at temperatures between 107°C and 163°C (225°F–325°F) comparing mercury capture on various sorbents in a packed bed under conditions where diffusion was not the limiting factor [29]. Several of the lignite-activated carbons tested were found to provide essentially 100% conversion of  $\text{Hg}^0$  to  $\text{Hg}^{+2}$  followed initially by complete capture, but the captured oxidized mercury

was not retained for extended periods of exposure to simulated flue gas. Results of a full-factorial matrix of experiments varying the gas species present determined that desorption of the oxidized mercury occurred when a combination of  $\text{SO}_2$  and  $\text{NO}_2$  was present, as will always occur to some degree in a coal-fired boiler. The mercury reemitted from the carbon sorbent has been shown to be a hydrate of mercuric nitrate [34]. Understanding and resolving this limitation will be essential to developing an effective carbon injection control method for mercury.

Control of nonvolatile toxic trace element emissions from coal combustion depends on achieving a high collection efficiency for fine particulates. In 1997, EPA issued a new ambient particulate matter standard limiting the concentration of the  $< 2.5\text{-}\mu\text{m}$  size fraction ( $\text{PM}_{2.5}$ ). In most regions of the United States, the  $\text{PM}_{2.5}$  in the atmosphere is derived predominantly from secondary sulfates and nitrates formed from  $\text{SO}_2$  and  $\text{NO}_x$  emissions. However, improved particulate control may be required on some combustion sources to meet the ambient standard, particularly on waste incinerators. An advanced hybrid particulate collector (AHPC) developed at the EERC achieves ultrahigh collection efficiencies as high as 99.9999% by synergistically combining membrane filtration and electrostatic precipitation in a unique geometric arrangement [31]. The AHPC offers superior particulate control in a potentially more economical design, and it has been licensed to W.L. Gore and Associates, for worldwide marketing.

## 9. Conclusion

Advances in combustion technology will be adopted only when they reduce cost and can be implemented with acceptable technical risk. Apart from technical risk, future decisions on new power plants will be principally influenced by trends in fuel cost, the efficiency and capital cost of new generating technologies, and environmental and regulatory policies including possible carbon taxes.

Referring to Table 1, conventional pc-fired plants burning coal at current price levels are competitive with GT combined-cycle plants burning natural gas priced at \$5/mscf (\$5/MMBtu). In the United States, between 81% and 85% of new generating capacity installed between 1997 and 2020 is projected to use natural gas [55]. US low-rank coals (lignite and subbituminous) that are used close to where they are mined typically offer a lower generating cost than most bituminous coals, depending on the value of a low sulfur content. Biomass gathered and delivered at a cost of \$40/dry short ton is not competitive with coal, but wood waste costing under \$1/MMBtu (ca. \$14/dry short ton) could be economical, depending on retrofit costs and the value of  $\text{SO}_2$  and  $\text{NO}_x$  credits.

The economic advantage of supercritical boilers shown in Table 1, assuming a capital cost \$100/kW higher than for a conventional pc-fired plant, is marginal at current coal prices in the United States. The small savings shown for high-cost bituminous coals is canceled for low-cost lignite, for which the value of increased efficiency is about equally offset by the higher capital cost. The advantage of a supercritical boiler for lignite is also diminished by the stack loss associated with high moisture content, which can be reduced only by drying the coal or lowering the gas temperature exiting the boiler. Any

economic factor that increases the cost of the fuel, including future carbon taxes, will improve the economics of supercritical boilers.

Future capital costs for advanced coal-fired power systems other than supercritical boilers are projected to be reduced to under \$900/kW as designs are simplified, and efficiencies are expected to increase to 45–50% as gas turbine inlet temperatures increase. Some designs, such as the hybrid gasifier–PFBC systems discussed in this report, may experience only a slight drop in efficiency for high-moisture coals owing to the increased mass flow through the gas turbine, which will especially benefit lignite-fired units. The cost of electricity for the coals in Table 1 at 45% HHV efficiency and \$900/kW capital cost is below the generating cost for combined-cycle natural gas at 50% HHV efficiency. Where large gas supplies are available, their generally lower extraction cost will allow gas to meet competition from any foreseen efficiency gain for coal. However, where gas is in short supply, prices will escalate and the stable price of coal will be an advantage.

The HiPPS design described in this paper, which heats air as the working fluid for a gas turbine, provides a conceptually simple way of combining the use of natural gas and coal to achieve very high efficiencies at projected capital costs between those for a supercritical boiler and a hybrid gasifier–PFBC design. Since the moisture in the coal does not pass through the gas turbine, lignite-fired HiPPS units will suffer about the same moisture-related losses as for a supercritical boiler. The HiPPS technology can be applied in either a new plant or in repowering an older plant, and it is closer to conventional pc-fired design than some competing systems.

Cofiring biomass with coal in pc-fired or cyclone boilers is fully feasible today, although some technical problems exist in regard to corrosion and deposition due to highly alkaline ash, fouling of SCR catalysts, and production of fly ash that does not meet current specifications for cement replacement. These problems will differ widely depending on the interactive properties of many different types of biomass and coal, and supporting research is needed to achieve trouble-free operation. With the exception of wood wastes used close to their source of supply, the cost of biomass is significantly higher than the current cost of coal. In addition, advanced power systems do not appreciably improve the economics of biomass relative to coal. Widespread adoption of cofiring will likely not occur in the United States without policy incentives.

The possible future adoption of a substantial tax on carbon emissions would change the relative economics of all of the fuels discussed here. A \$50/ton carbon tax would level the playing field and bring costs for biomass closely into line with coal. Natural gas is relatively less affected by a carbon tax compared to coal owing to its lower carbon content in relation to calorific value. After factoring in the \$50/ton carbon tax in Table 1, the generating cost for natural gas at \$5/mscf remains competitive with conventional technology, not with high-efficiency systems firing either coal or biomass that is exempted from the tax.

Low-cost high-efficiency power systems for coal still face technical challenges, which are delaying their commercial deployment. For the HiPPS design described in this paper, the technical barriers concern the development of more corrosion-resistant alloy and ceramic materials for extreme high-temperature service. For the hybrid (topping) gasifier–PFBC systems, the barriers concern the cost and reliability of hot-gas filters for

removing fine particles that would erode turbine blades. Supercritical boilers are being built today, but their application to high-alkali lignites involves poorly understood technical risks due to high-temperature fireside corrosion. All of these advanced coal technologies, as well as conventional designs, may face additional challenges from new environmental regulations concerned with mercury and other toxic trace element emissions, for which controls remain to be fully developed. Research and development is continuing on all of these barrier issues, and success is within reach over the next decade. Coal will of necessity continue to be used in many regions that do not have acceptable economic alternatives. The current authors believe that the specter of global warming which is cooling the prospects for coal will be overcome, partly through the efficiency gains, and eventually by the sequestering of the carbon dioxide that is produced. Coal is part of the future.

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