

The Removal of Impurities from Hot Coal-Derived Gas by Filtration

by

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At the Morgantown Energy Technology Center (METC) of the U.S. Department of Energy, an effort is underway to clean hot coal-derived gas to a sufficient purity for combustion in heat engines and other applications. Experiments were conducted in which the hot product gas, raw and desulfurized, from a fluid-bed coal gasifier was filtered at temperatures up to 1,600°F, pressures up to 250 psig, and flow rates up to 2,500 scfh. Particulate and condensate samples were collected at the inlet and outlet of the filters and analyzed for 10 metals which are common constituents of coal ash. The results were used to determine how filtration of particles from a gas stream at high temperatures affected metal concentrations in the gas. In general, the removal efficiency for the metals correlated with the particulate removal efficiency.

Introduction

Under the Advanced Environmental Control Technology Program established in 1979 by the Department of Energy (1), Morgantown Energy Technology Center has been actively engaged in the development of technology necessary to control contaminants from coal gasification that are deleterious to gas turbines and hazardous to the environment. A significant part of this effort has been concerned with particulate and trace metal, particularly alkali metals, removal from the hot gas streams. Three hot gas filtration devices developed in the program by contractors were tested at METC to determine their performance in an actual hot gas stream produced by a small-scale, fluid-bed gasifier (2). The three devices were a Westinghouse ceramic cross-flow filter, an Acurex ceramic bag filter, and an electrostatic precipitator developed by Denver Research Institute. Details of the design of these devices and results of the work to date have been reported in a series of contractor review meetings (3-9).

Although the principal purpose of this series of tests was to measure the particulate removal efficiency of the hot gas filters at temperatures up to 1,850°F, they afforded the opportunity to also measure the concentration of 10 metals, which are common constituents of coal ash, in the gas stream. Specifically, it was desired to determine whether removal of particles from a coal gas at temperatures up to 1,850°F would affect the downstream concentration of trace species of metals such as sodium and potassium. The results of the experiment are reported in this paper.

Experimental

The low-Btu coal gas used during these tests was produced by METC's advanced concepts fluid-bed gasifier. The gasifier was designed to operate with a wide range of coals at pressures of 200 to 1,000 psig and temperatures up to 1,900°F. The gasifier is used as a source of low- to medium-Btu gas for use in testing downstream components and analytical instrumentation.

During these tests, the gasifier was operated at 400 psig with Montana Rosebud subbituminous coal, air, and steam to produce a gas of the following average composition:

N ₂ :	38.0 Percent	CO ₂ :	10.0 Percent
H ₂ O:	17.0 Percent	CH ₄ :	3.5 Percent
H ₂ :	17.0 Percent	H ₂ S:	0.3 Percent
CO:	13.5 Percent		

Approximately 300 feet of insulated pipe separated the gasifier from the cleanup device test area. Heat loss and particle dropout in the pipe resulted in a gas temperature and solids loading that were lower than that required for the tests. Thus, a methane-fired heater was used to increase the temperature to the desired range (1,400° to 1,850°F), and a pressurized, screw-type particle feeder was used to feed particles into the gas stream up stream of the test device. These particles were obtained from a cyclone on the gasifier and had the same composition as the entrained particulate. The rate of particulate addition was varied depending on the amount already in the gas and the requirements of each test. Typically, the added particles were approximately 10 to 40 percent of the total at the inlet of the test devices. After exiting the cleanup device, the gas was cooled to remove condensibles, metered, and then incinerated.

Tests conducted with the Westinghouse and Acurex filters lasted 250 hours each. The test of the DRI electrostatic precipitator was terminated after approximately 50 hours because of a mechanical failure. During 50 hours of the Acurex filter test, the coal gas was desulfurized prior to particulate removal. This was accomplished using METC's hot gas desulfurization unit which uses a zinc ferrite (ZnFe₂O₄) sorbent to remove gaseous sulfur species. This device typically removes greater than 99 percent of the sulfur in the gas and produces gas streams with sulfur concentrations of less than 10 ppm (8). The configuration of the various components of the test system are shown in Figure 1.

Particulate sampling probes were positioned at various points in the system to characterize the performance of each device. To measure the solids loading of the gas produced by the gasifier, a sampling probe was placed in the gas line after the primary cyclone. Sampling probes were installed before and after the hot gas desulfurization unit to determine if this device removed particles from the gas stream or if zinc ferrite bed material was being lost by attrition.

The data reported here were collected at the inlet and outlet of the particulate removal test devices. The inlet probe was positioned downstream of the particle feeder, but upstream of the test device. The outlet sampling system was designed to filter the total system flow exiting the test device.

Sampling System Design

The inlet sampling probe was constructed of 1/4-inch stainless steel tubing with either a 0.035- or 0.049-inch wall thickness. The tip of the probe was beveled approximately 1/4 inch. When the probe was operated isokinetically, approximately 3 to 5 percent of the total process flow was sampled. The probe was positioned in a process pipe elbow with the probe tip approximately 6 inches upstream of the start of the bend.

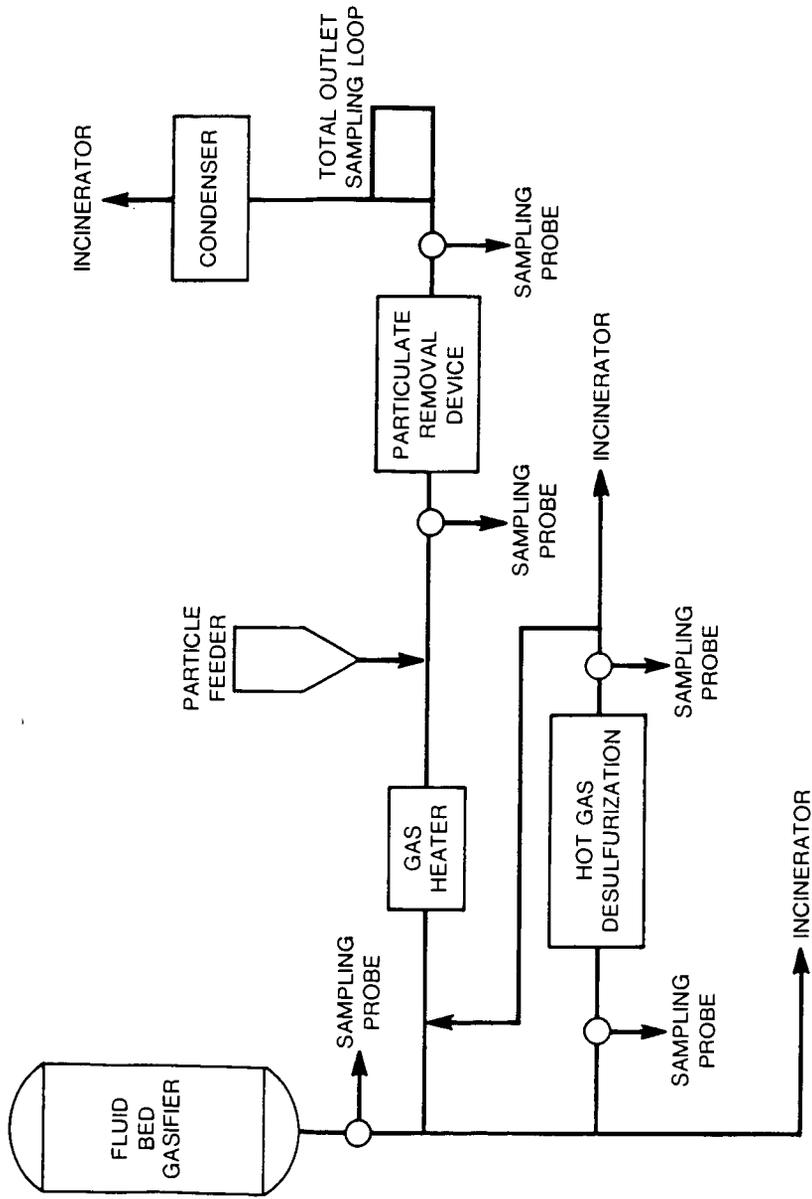


Figure 1
System Components

Figure 2 shows the inlet sample line configuration. The particulate samples were collected using 1.37- by 3.94-inch (35- by 100-millimeter), rounded bottom, alundum ceramic extraction thimbles (Norton No. 6406). The thimbles are designed to withstand temperatures up to 2,600°F, and pressure drops of up to 100 psi. The entire sampling line and gate valves were wrapped in high-temperature heating tapes, insulation, and a high-temperature glass cloth (Colombia Fiberglass). The heating tapes, controlled by digital temperature controllers (Omega Model No. CS4001KF), were maintained at 1,000°F. It was necessary to use an alternative heating method for the thimble holders because of their high mass. An 8-foot by 1/8-inch heating rod (ARI Industries, Inc.) was coiled around the elongated section of a thimble holder by means of metal hose clamps. The heating rods were powered from variable autotransformers set at 100 to 120 volts. The gas temperature at the downstream end of the thimble holders was typically 350° to 400°F for the inlet samples and 700° to 800°F for the outlet samples.

Approximately 30 feet of 3/8-inch stainless steel tubing separated the thimble holder from the flow monitoring system. This was necessary because of the shortage of available space in the test device area. The flow monitoring system consisted of a cooling coil, a condensate trap, and a flowmeter (rotameter). Condensate samples were collected in a 1-liter, stainless steel sampling cylinder (Hoke No. 8LD1000).

The flowmeter, a temperature indicator and a pressure gauge were located between two 1/4-inch needle valves. By adjusting these valves, the desired flow could be maintained at a known pressure and temperature. After metering, the gas was exhausted to an incinerator.

To take a particulate sample, the probe, gate valves, and thimble holder were first heated for 1 hour. Just prior to starting sample collection, the probe was purged with nitrogen for 3 to 5 seconds. The gate valves were opened and then the needle valves on the flow monitoring system were adjusted to set the sample gas flow rate. Sampling times were typically 1 to 2 hours.

After sample collection was completed, the thimble holders were disconnected and allowed to cool before removing the loaded thimble. The condensate was drained and measured so that the contribution of water vapor to the total gas flow could be calculated. The 30-foot section of tubing between the thimble holder and the flow monitoring system was purged with steam and nitrogen after each sample to remove residual condensate from the tubing walls. This was added to the condensate sample.

Figure 3 depicts the configuration of the total outlet sampling system. The sampling system was designed as a bypass loop from the main gas line. The entire gas flow was filtered using two thimble holders in parallel and then the gas was cooled to condense water vapor. The sample loop was isolated by gate valves (Vogt No. 821) upstream and downstream of the thimble holders. The sample loop and thimble holders were insulated and heated as described previously. The condensate was collected in stainless steel vessels which were drained before and just after each outlet sampling. The weight of the condensates was recorded so that the contribution of water vapor to the total gas flow could be calculated. Metal concentrations in the condensate could also be translated into equivalent total gas concentrations.

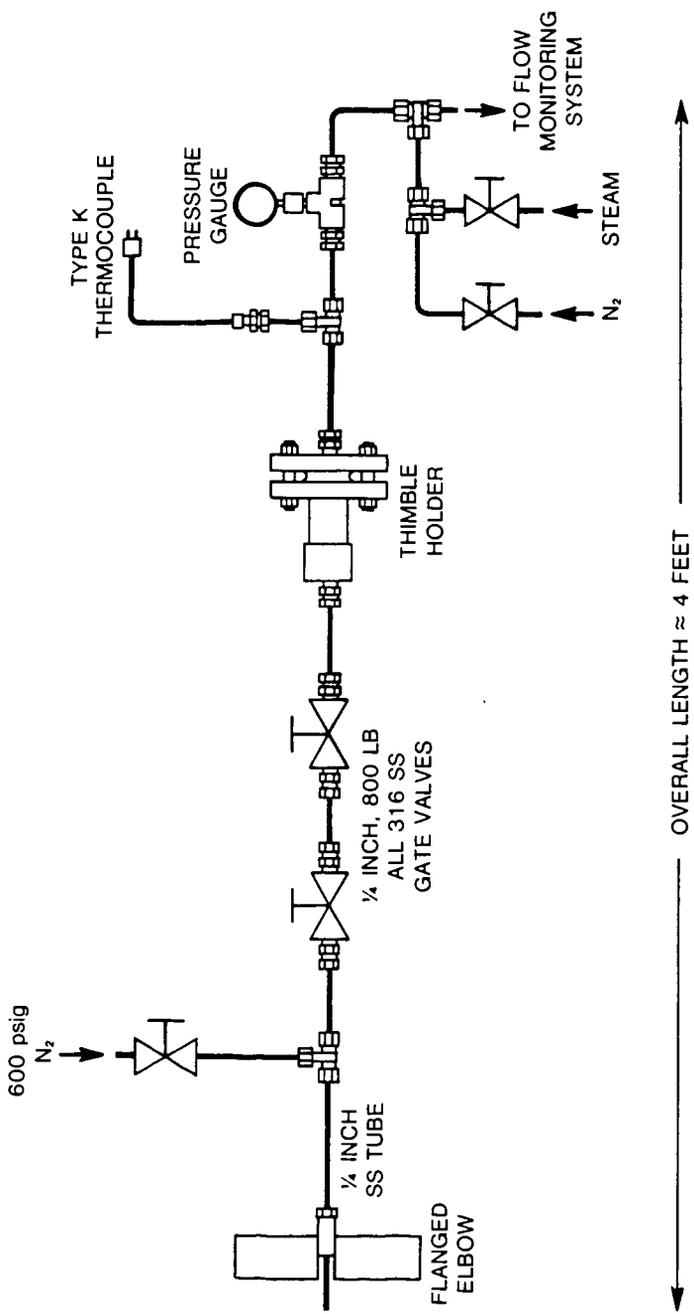


Figure 2
Sampling Probe Configuration

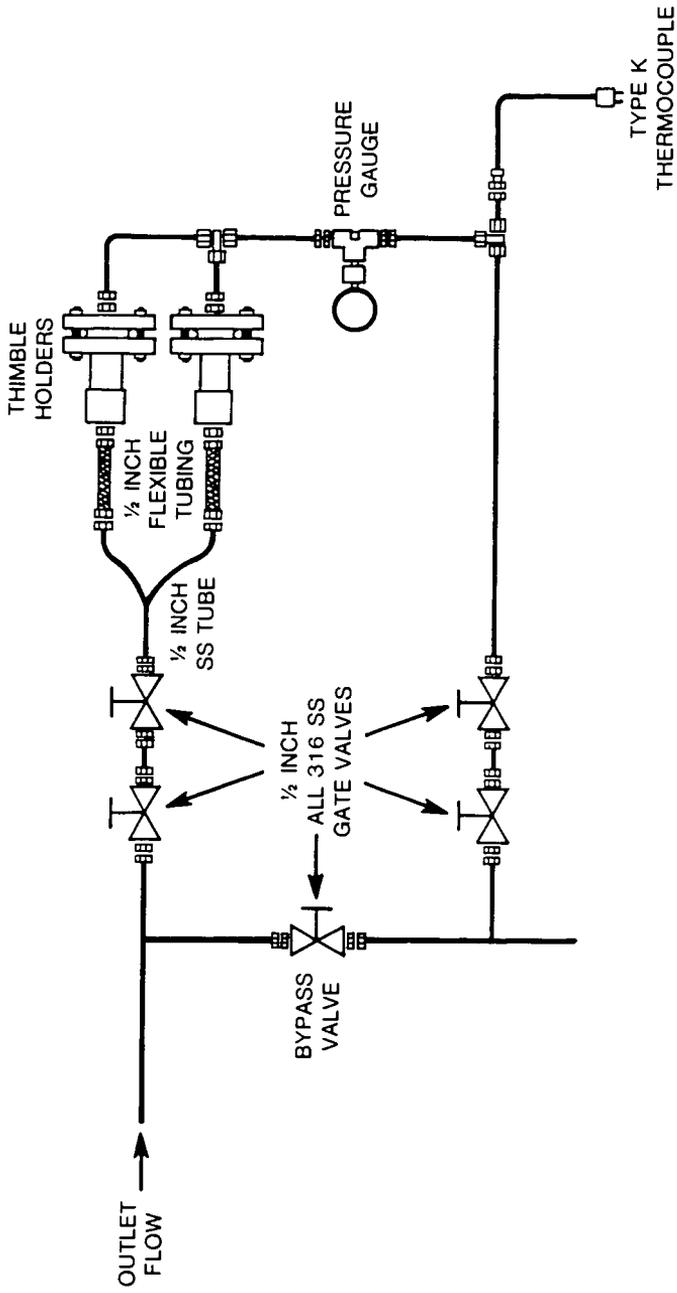


Figure 3
Total Outlet Sampling Configuration

Sample Preparation

Thimbles were heated in an oven at 250°F for approximately 30 minutes, cooled in a desiccator, and weighed on an analytical balance prior to being loaded into a thimble holder. After collecting a particulate sample, the thimble holders were extremely hot and were allowed to cool for approximately 30 minutes. The holder was then taken apart and the thimble removed. The thimble, containing the particulate sample, was placed in an oven at 250°F for approximately 45 minutes. After cooling, the thimble was weighed again. The difference in the initial and final weights was taken to be the weight of particles on the thimble. Typically, a small fraction of the particles escaped around the edge of a thimble while in the holder. These particles were collected together with the condensates and filtered out using Gelman Metricel, 47-millimeter, 0.45-micron, Type GA-6 filters under vacuum. The sample was then dried in an oven, cooled in a desiccator, and weighed. This weight was added to the weight of particles collected on the thimble to yield a total sample weight. Portions of the particles collected in the thimbles were submitted for particle size, ultimate, and metals analyses. Particles filtered from the condensates were not used for the chemical analyses because of the possibility that leaching had affected their composition. Particle size analyses were performed using a Model No. TALL Coulter Counter. The pH of the condensates was adjusted to ≤ 2.0 by addition of nitric acid, and stored in polyethylene bottles.

The solid and condensate samples were analyzed for 10 metals using a Perkin-Elmer Model 5000 atomic absorption spectrophotometer and a Spectrametrics, Inc., direct plasma atomic emission spectrometer. A lithium metaborate fusion was performed on the solid samples in order to generate a liquid for these analyses.

Data Analysis

As described in the experimental section, the concentrations of the metals in the gas stream were obtained by measuring their amounts in the particles and condensate collected utilizing sampling devices at the inlet and outlet of the test devices, and relating them to the quantity of gas which had flowed through the devices. In the course of the tests, 18 sets of inlet/outlet samples were taken for the Westinghouse ceramic cross-flow filter, 23 for the Acurex fabric bag filter, and 10 for the Denver Research Institute electrostatic precipitator.

The performance of the 3 filters are summarized in Table 1, which gives the average particulate loading of the inlet and outlet gas together with the calculated removal efficiency and the mean particle diameter. The average ultimate analyses of the particulate samples are detailed in Table 2, and Table 3 gives the average gas concentrations of particles, ash, and 10 metals. The calculated values of removal efficiency are also included.

Generally, the Westinghouse filter had the highest particulate removal efficiency followed by the Acurex and DRI filters. Removal efficiency tended to increase with inlet particle loading. The Westinghouse filter tended to result in a lower outlet particle loading than the Acurex or DRI filters because of its higher particulate removal efficiency.

Statistical analysis of the data indicates that, though not strong, there is a correlation between particulate removal efficiency and each of the metal removal efficiencies. The metal removal efficiencies significantly are some

2 to 6 percentage points lower, on average, than the particulate removal efficiency. This cannot be accounted for by gasification of carbon in the particulate on passing through the filters, for a similar result is obtained when compared with the ash removal efficiencies. This may be due to a higher metals concentration in the smaller particles passing through the filters.

A particularly interesting result is that metal removal efficiencies could not be shown to be dependent on operating conditions which varied in the ranges: temperature -- 800° to 1,600°F; pressure -- 140 to 250 psig; and flow rate -- 1,300 to 2,500 scfh. This is especially important for the case of the alkali metals, sodium and potassium, since it is speculated that, at higher temperatures and lower pressures, these relatively volatile species might pass more readily through the hot gas filters.

The average measured removal efficiency for the metals varied slightly among themselves. This is shown in Table 4. In Table 5, the metal concentrations are expressed as the average percentage of the total sum of the ten metals for both the inlet and outlet. These data show that there was an enrichment of Ca in the outlet gas. This was most apparent during the Westinghouse test during which the average Ca concentration increased from 20 percent of the total of the metals at the inlet to 28 percent of the total at the outlet. The source of this enrichment was believed to be the refractory lining of the piping. This assertion is supported by the fact that after the Westinghouse test, the original refractory was replaced with a new refractory that was rated for a higher temperature. Subsequently, during the Acurex and DRI tests Ca enrichment of the outlet gas continued but to a lesser degree. The outlet gas was enriched in Mg and alkalis as well, but to a lesser degree than Ca. These metals are also components of the refractory. The composition of the two refractories is shown in Table 6.

During 50 hours of the Acurex filter test, the hot gas to the filter was first desulfurized by passage through a fixed bed of zinc ferrite (8). The resulting gas tended to have a lower particulate loading comprising smaller size particles. No particulate was added to the gas stream during this period. However, the particulate removal efficiency during this period did not differ significantly from test periods when raw gas was utilized. Nor was it found that the particulate was enriched in zinc, which might have been a consequence of attrition or vaporization of the zinc ferrite bed. The only metal which showed a significant difference in removal efficiency during the test using desulfurized gas was nickel. The efficiency during the desulfurization test was lower.

TABLE 1. CHARACTERIZATION OF PARTICLES IN THE HOT COAL GAS STREAM

	Average Inlet Loading (mg/scf)	Average Outlet Loading (mg/scf)	Average Particulate Removal Efficiency %	Standard Deviation Removal Efficiency %	Average Inlet Mean Diameter (micron)	Average Outlet Mean Diameter (micron)
WESTINGHOUSE (18 samples)	65.91	0.86	98.15	1.42	9.76*	7.50**
ACUREX (23 samples)	64.38	1.91	95.59	4.38	8.76	7.00
DENVER RESEARCH INSTITUTE (10 samples)	72.36	5.03	93.29	4.25	8.84	8.70

* Inlet particulate size data available for 11 samples.

** Outlet particulate size data available for 10 samples.

TABLE 2. ULTIMATE ANALYSIS OF PARTICULATE SAMPLES

Filter	Location	Average Weight Percent				
		Ash	C	H	N	S
ACCUREX	Inlet	34.34	60.12	0.49	0.51	0.83
	Outlet	37.63	59.59	0.37	0.42	0.69
DRI	Inlet	39.34	55.10	0.38	0.69	1.20
	Outlet	38.18	56.94	0.30	0.69	0.85
WESTINGHOUSE	Inlet	34.61	58.22	0.53	0.88	1.36
	Outlet	39.94	55.47	0.44	0.87	1.36

TABLE 3. REMOVAL OF PARTICLES AND METALS FROM HOT COAL GAS

		Average Inlet Concentration (ppm)	Average Outlet Concentration (ppm)	Removal Efficiency (%)
WESTINGHOUSE	Particles	2,365.37	30.87	98.7
	Ash	742.00	12.40	98.3
	Al	105.27	1.75	98.3
	Ca	58.93	1.89	96.8
	Cr	2.62	0.07	97.3
	Cu	1.40	0.04	97.1
	Fe	91.09	2.29	97.5
	K	8.85	0.25	97.2
	Mg	21.08	0.46	97.8
	Na	4.62	0.12	97.4
	Ni	2.44	0.08	96.7
	Zn	0.84	0.03	96.4
	ACUREX	Particles	2,310.50	68.55
Ash		718.00	23.80	96.7
Al		89.99	3.09	96.6
Ca		48.51	2.40	95.1
Cr		1.08	0.06	94.4
Cu		1.53	0.04	97.4
Fe		76.11	2.82	96.3
K		8.89	0.47	94.7
Mg		18.22	1.09	94.0
Na		3.06	0.22	92.8
Ni		0.61	0.05	91.8
Zn		0.60	0.04	93.3
DRI		Particles	2,596.90	180.55
	Ash	969.00	57.7	94.0
	Al	173.61	23.59	86.4
	Ca	46.36	5.88	87.3
	Cr	0.92	0.10	89.1
	Cu	0.73	0.08	89.0
	Fe	113.89	12.80	88.8
	K	21.40	2.12	90.1
	Mg	15.50	2.49	83.9
	Na	5.75	0.72	87.5
	Ni	3.53	0.60	83.0
	Zn	0.62	0.06	90.3

TABLE 4
 AVERAGE REMOVAL EFFICIENCIES FOR
 EACH METAL IN ORDER OF DECREASING
 EFFICIENCY

Metal	Number of Samples	Average Efficiency (%)
Al	50	93.89
Fe	51	93.73
Cr	51	93.21
K	51	92.83
Cu	51	92.37
Zn	51	91.89
Na	51	91.56
Ca	51	91.05
Mg	51	90.92
Ni	51	89.52

TABLE 5. AVERAGE INLET AND OUTLET PERCENT METAL COMPOSITIONS

Metal	Location	Average Metal Composition (% of Total of Ten Metals)			
		Acurex	DRI	Westinghouse	Average
Al	Inlet	35.50	46.16	32.30	36.46
	Outlet	27.73	42.72	25.22	29.78
Ca	Inlet	22.01	12.88	20.02	19.52
	Outlet	25.56	15.51	28.09	24.48
Cr	Inlet	0.64	0.26	1.45	0.85
	Outlet	0.87	0.47	1.15	0.89
Cu	Inlet	1.14	0.21	0.69	0.79
	Outlet	0.62	0.63	0.57	0.60
Fe	Inlet	27.18	28.15	31.78	28.99
	Outlet	25.43	23.10	30.14	26.64
K	Inlet	3.22	5.13	3.01	3.52
	Outlet	4.12	4.81	4.01	4.22
Mg	Inlet	8.19	4.34	7.39	7.15
	Outlet	10.24	8.49	7.07	8.78
Na	Inlet	1.28	1.49	1.69	1.47
	Outlet	4.20	1.92	1.91	2.94
Ni	Inlet	0.41	1.21	1.33	0.89
	Outlet	0.70	1.95	1.35	1.18
Zn	Inlet	0.44	0.18	0.35	0.36
	Outlet	0.52	0.41	0.50	0.49

TABLE 6. PIPELINE REFRACTORY COMPOSITIONS*

Test	Al ₂ O ₃ %	SiO ₂ %	Fe ₂ O ₃ %	TiO ₂ %	CaO %	MgO %	Alkalis %
Westinghouse ¹	13.95	30.29	5.34	0.14	35.39	8.81	2.92
Acurex, DRI ²	38.61	31.48	5.38	1.50	19.54	0.79	1.41

* Plibrico Company Laboratory Report.

¹ Refractory maximum temperature rating was 1,500°F.

² Refractory maximum temperature rating was 2,000°F.

Conclusions

Analysis of inlet and outlet particulate samples in a hot coal gas stream passing through three different filtration devices indicates that 10 metals, commonly occurring in coal ash, were removed from the gas stream in proportion to the particulate removal. This remained true up to a temperature of 1,600°F, even for relatively volatile metals such as sodium and potassium. Metal removal efficiencies could not be shown to be dependent on operating conditions for temperatures up to 1,600°F, pressures up to 250 psig, and flow rates up to 2,500 scfh. The efficiency of removal of the metals was slightly less than the particulate removal efficiency, suggesting an enrichment of the metals in the smaller particles escaping through the filter.

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