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of Mercury, Arsenic, and Selenium from Coal  
by Dry Magnetic Separation***

Appearing in the Proceedings of the 24<sup>th</sup> International Technical  
Conference on Coal Utilization and Fuel Systems  
Clearwater, FL, March 8-11, 1999  
(CSTA, Washington, DC) pp. 151-158

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## **Preliminary Results of Pre-combustion Removal of Mercury, Arsenic, and Selenium from Coal by Dry Magnetic Separation**

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We report the results of preliminary measurements of pre-combustion separation of major metal oxides and trace elements from coal by dry magnetic separation. The measurements have been made as part of ETCi's development of MagMill™ technology for removing mineral matter from coal at the pulverizer at the front end of a coal fired power plant. The technology is specific to separation of mercury, arsenic, and selenium because of their associations with iron pyrites in coal. Measurements were made on a suite of five Eastern U.S. and five Illinois Basin bituminous rank coals prepared at 8 Mesh topsize and processed as 8 Mesh by zero fractions through a dry ParaTrap Magnetic Separator. Measurements of major metals and trace elements were made on the feed coal, the magnetic refuse fraction and the magnetic clean coal product. The range of weight recoveries measured for 13 of the trace elements for the suite of coals indicates a significant potential for pre-combustion removal of trace elements and especially for mercury, selenium, and arsenic by dry magnetic methods. While these three elements are important because they are considered hazardous air pollutant precursors, pre-combustion removal of arsenic is especially important because of its role in poisoning catalysts used in emerging SO<sub>x</sub> and NO<sub>x</sub> control technologies.

### **I. INTRODUCTION**

There are eleven trace elements (Sb, As, Be, Cd, Cr, Co, Pb, Mn, Hg, Ni, and Se) in the list of 189 Hazardous Air Pollutants (HAPs) identified in Title III of the Clean Air Act Amendments (CAAA) of 1990. Work sponsored by DOE<sup>1</sup> indicates that emission of the nonvolatile trace elements identified as HAPs (Sb, As, Be, Cd, Cr, Co, Pb, and Mn) can be effectively controlled (>98%) by highly efficient post combustion particulate control technology or technology combinations<sup>2</sup> currently being used by the U.S. utility industry.

Mercury and selenium are the exceptions. For selenium it is concluded that with available information "...it is not possible to clearly delineate the potential to control Se emissions from coal-fired boilers using existing emission control technologies."<sup>3</sup> The report to

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<sup>1</sup> "A Comprehensive Assessment of Toxic Emissions from Coal-Fired Power Plants, Phase I Results from the U.S. Department of Energy Study" prepared for: Pittsburgh Energy Technology Center, Morgantown Energy Technology Center by: Energy & Environmental Research Center, University of North Dakota, P.O. Box 9018, Grand Forks, North Dakota 58202-9018, September, 1996.

<sup>2</sup> Electrostatic Precipitator-Flue Gas Desulfurization, or Spray Drier Absorber-Fabric Filters.

<sup>3</sup> Reference 1, op cit.

the DOE further states "...the potential to control Hg emissions from coal-fired utility boilers using existing emission control technologies is unclear." This is because mercury is volatile and its control in the vapor phase will not likely exceed that of other priority pollutants such as SO<sub>2</sub> and because of inadequacies in the speciation of mercury in current analytical practice. The DOE concludes "... existing control technologies will require augmentation, and alternative control technology options will require development."

Conventional physical coal cleaning technology is one option. It separates some trace metals which associate with gross mineral impurities. It is now widely practiced in the U.S. where about 75% of the coal produced east of the Mississippi River which is burned by utilities is cleaned in coal preparation plants. This approach to physical separation of trace metals prior to burning is of limited utility, however, because of the use of water in cleaning coal at the mine. It is impractical to wet clean coal at particle sizes which free the minerals and trace metals so that they may be separated efficiently.<sup>4</sup>

The use of dry technology offers an attractive new option in cleaning coal which avoids the limitations incurred in using water. It is not now widely practiced, however, because until recently there has not been a practical method for dry cleaning fine coal. The MagMill™<sup>5</sup> is a **new** concept for dry cleaning pulverized coal at the point of use --- which may be pulverized coal-fired power plants, blast furnaces, cement kilns, or other large scale coal users. A MagMill™ is a dry magnetic separator attached to or retrofitted to a pulverizer. It is used to separate mineral gangue as it is liberated from the coal matrix inside the pulverizer. Coal cleaning with a MagMill™ is more efficient and less expensive than conventional wet cleaning because the coal is cleaned in much finer size consists than is practical at the mine. In most utility applications the pulverizer is already in use and need not be charged to the cleaning process. Clean coal can be prepared and trace element carryover to the burner can be minimized using the MagMill™ technology which **lowers** rather than increases fuel cost!

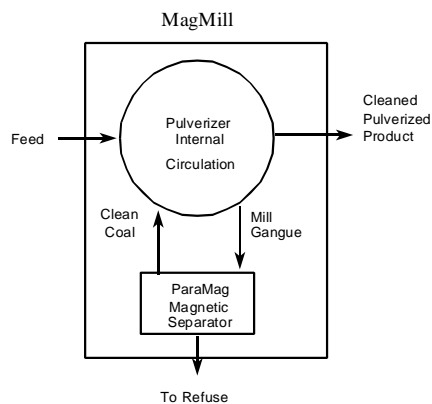


Fig. 1. MagMill™ Concept

The MagMill™ technology is specific to removal of iron pyrite and other hard minerals. It is known that selected species of trace metals in coal, namely mercury and arsenic, associate with the iron pyrites and can be separated with them<sup>6</sup>. Preliminary survey measurements at ETCi

<sup>4</sup> Gerald H. Luttrell, Gregory T. Adel, and Roe-Hoan Yoon, "Precombustion Removal of Hazardous Air Pollutant Precursors." Proc. of the Coal Liquefaction & Solid Fuels Contractors Review Conference, Pittsburgh, PA Sept. 3-4, 1997, DOE/FETC/PGH-97001.)

<sup>5</sup> Y. Feng, et al., "Dry Coal Cleaning in a MagMill," Proc. of the 22<sup>nd</sup> International Technical Conference on Coal Utilization and Fuel Systems, Clearwater, FL, March 16-19, 1997.

<sup>6</sup> See the Chemistry of Coal Utilization, Prepared for the Commission on Natural Resources, National Research Council, National Academy of Sciences, National Academy of Engineering, Wiley-Interscience. See also Robert B. Finkelman, "The Use of Modes of Occurrence Information to Predict the Removal of the HAPs Prior to Combustion," J. Coal Qual. 12, n. 4, p. 132(3), (Oct-Dec, 1993).

which are reported here have shown that unoptimized dry magnetic separation alone can reduce trace elements in coal, and especially mercury and arsenic, to levels obtained by conventional coal cleaning. Marriage of the magnetic separator and the pulverizer to make a MagMill™ offers a unique opportunity to develop an innovative technology which will mitigate emissions of HAPs while also lowering fuel costs for the utility. Also, pre-combustion removal of arsenic can help alleviate problems of catalyst poisoning in selective removal of post-combustion nitrogen oxides.

## II. EXPLORATORY MAGNETIC SEPARATION

Trace elements were measured for the feed, clean coal, and magnetic separator refuse fractions for ten coals supplied by an electric utility. Trace element measurements for beryllium, copper, arsenic, selenium, and/or mercury in the magnetic fractions for three of the coals resulted in calculated recoveries in excess of 150% of the element in the feed coal. Since the source of error could not be traced, these coals were excluded from the analysis given here. The results of dry magnetic separation measurements are reported for the remaining seven (7) raw coals which did not exhibit extreme unbalance in the recovery of the trace elements. The coals are Appalachian and Illinois basin U.S. bituminous rank raw coals. Their states of origin and major characteristics are shown in Table I.

Table I  
State of Origin and Major Characteristics of Raw Coals Studied

Coal No.	State of Origin	Ash (Wt.%)	Sulfur (Wt.%)
1	SW Virginia	12.22	1.97
2	N West Virginia	38.79	3.29
3	SW Pennsylvania	17.62	2.86
4	W Kentucky	15.22	1.84
5	W Kentucky	18.71	4.62
6	W Kentucky	20.76	4.56
7	S Illinois	26.20	4.23

The coals were ground to 8 mesh topsize and processed as 8 mesh by zero through a ParaTrap dry magnetic separator in a screening study to investigate the feasibility of separating minerals from these coals. The feed, refuse, and clean coal products from the separator were ashed, and the ash fractions were analyzed for major metals and trace elements by the electric utility using ASTM procedure D3684. ASTM D3684 employs an oxygen bomb to combust the sample in a pure oxygen atmosphere. A small amount of acid is placed in the bomb to “trap” the mercury as it is released.

## III. RESULTS

Tables II-IV show the analysis for the feed, magnetic separator refuse, and magnetic separator clean coal fractions for the coal from southwestern Virginia. This result is typical of all of the measurements.

Table II  
Proximate and Ultimate Analysis of Magnetic Fractions  
for Southwestern Virginia Coal

Item	Feed (Wt.% Coal)	Clean (Wt.% Coal)	Refuse (Wt.% Coal)
Recovery	100.00	87.74	12.26
Ash	12.22	8.16	41.94
Moisture	2.49	2.46	2.56
Volatile Matter	35.24	36.70	25.91
Fixed Carbon	52.43	55.15	32.14
Heating Value	13094	13770	8039
Carbon	75.82	79.38	45.73
Hydrogen	4.82	5.09	2.74
Nitrogen	1.44	1.49	0.82
Oxygen	3.75	4.33	3.99
Sulfur	1.95	1.55	4.77
Chlorine	0.05	0.05	0.02

For this coal, single pass, unoptimized dry magnetic separation reduced the ash of the raw coal by 33% and the sulfur by 21% with an 87.74% weight recovery.

Table III  
Analysis of Ash in Magnetic Fractions  
for Southwestern Virginia Coal

Item	Feed (Wt.% Ash)	Clean (Wt.% Ash)	Refuse (Wt.% Ash)
P <sub>2</sub> O <sub>5</sub>	0.33	0.44	0.22
SO <sub>3</sub>	1.15	1.68	0.54
SiO <sub>2</sub>	51.3	51.7	51.2
Fe <sub>2</sub> O <sub>3</sub>	13.1	10.9	16.6
MgO	1.09	0.89	1.35
TiO <sub>2</sub>	1.19	1.40	0.91
Al <sub>2</sub> O <sub>3</sub>	27.70	29.30	24.70
CaO	0.96	1.40	0.35
Na <sub>2</sub> O	0.24	0.24	0.23
K <sub>2</sub> O	2.86	2.05	3.79

Table IV  
Trace Metals in Magnetic Fractions of a Southwestern Virginia Coal

Element	Trace Element Concentration			Recovery
	Feed	Refuse	Clean	Clean
	(ppm Coal)	(ppm Coal)	(ppm Coal)	(ppm Coal)
P	112.91	179.08	100.37	77.99
Cr	15.64	39.42	12.73	71.41
Zn	15.12	41.52	12.57	71.05
Cd	1.83	7.13	1.14	54.68
Pb	12.59	33.13	11.83	82.48
Ni	13.81	27.26	12.65	80.37
Ba	114.26	252.90	92.21	70.81
Mn	12.46	46.13	7.59	53.42
V	27.13	66.27	22.03	71.26
Be	1.10	2.10	0.98	78.12
Cu	16.99	28.10	15.50	80.09
Hg	0.11	0.29	0.05	41.56
Se	5.13	0.4	4.24	72.54
As	4.77	6.29	4.32	79.62

The numbers in the right column of Table IV show the recoveries of the elements in the clean coal fractions. About 42% of the mercury, 73% of the selenium, and 80% of the arsenic is recovered in the clean coal fraction in these survey type measurements.

The range of recoveries for the major and minor elements in the magnetic separator clean coal fractions observed for the seven coals is shown in Figure 2 where the recoveries are based on feed coal. In the graph, the top of the bar is the highest recovery and the bottom is the lowest. The observed recoveries generally range from 30% to 90% with an exception for chlorine which exhibits an anomalous result. The weighted average of the recovery for each of the elements for the seven samples is shown as the horizontal line inside the bars. The average recoveries for elements other than chlorine are around 50%. Chlorine is higher at about 90%. Four of the coals are at or below detection limits for the method used for chlorine determination (ASTM 2361).

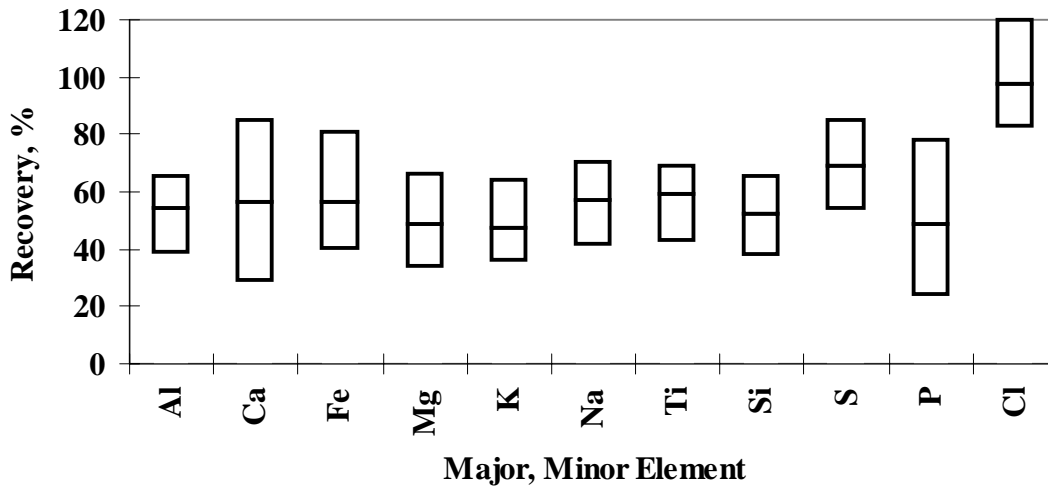
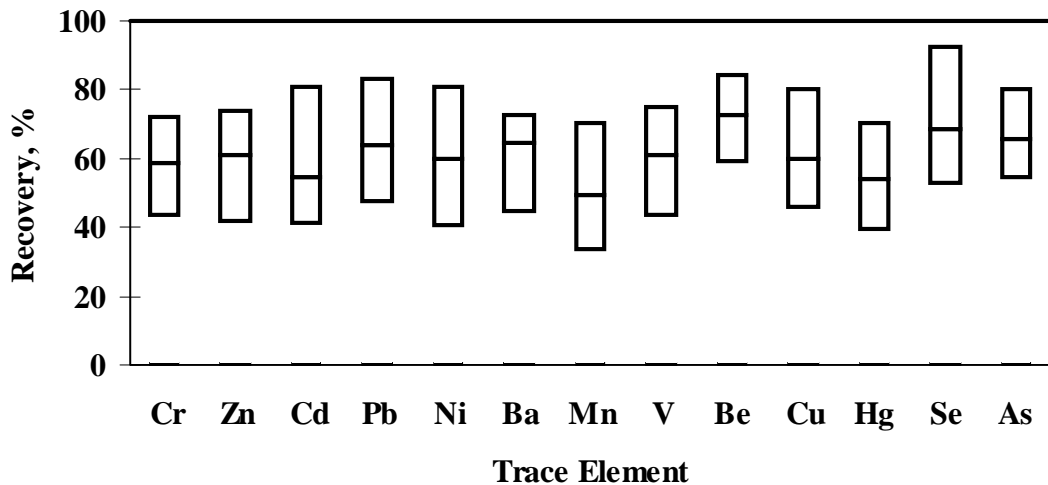


Fig. 2. Recovery of Major and Minor Elements in the Magnetic Separator Clean Coal Fractions for 7 U.S. Bituminous Rank Coals

The range of recoveries of trace elements observed for the seven coals is shown in Figure 3. Here, the top of the bar is the highest recovery and the bottom is the lowest. The weighted average of the recovery for each of the elements for the seven samples is shown as the horizontal line inside the bars. The averages are approximately 55%, 65%, and 60% for mercury, selenium, and arsenic respectively.

Fig. 3. Range of Recovery of Trace Elements



Concentrations of the trace elements have been correlated with the ash levels in the magnetic fractions for each coal. The average values of the coefficient of fit, Rsquare, and of the standard deviation of the Rsquare values for the seven measurements are given in Table V for each trace element. The concentration of each element correlates well with the ash level in the coal fractions in which it was measured. The goodness of fit, as measured by the average

Rsquare values and the standard deviation, decrease beginning with Be and are the poorest for As.

Table V  
Relation of Concentration of Elements in Seven Bituminous Coals with Ash Level  
Average “Goodness of Fit”

Trace Element	Goodness of Fit	
	(Avg RSq)	(Stdev)
P	0.9995	0.0007
Cr	0.9994	0.0008
Zn	0.9994	0.0010
Cd	0.9992	0.0017
Pb	0.9989	0.0023
Ni	0.9984	0.0016
Ba	0.9981	0.0030
Mn	0.9977	0.0030
V	0.9972	0.0042
Be	0.9916	0.0158
Cu	0.8987	0.1215
Hg	0.8657	0.3497
Se	0.8447	0.2189
As	0.8062	0.3330

Dry magnetic separation provides an easy, convenient, and rigorous method for studying the relationship of trace elements to the minerals in coal. Use of techniques such as X-Ray diffraction or electron microscopy or even forms of sulfur measurements, not made for this work, could focus the results by identification of mineral forms.

#### IV. FUTURE WORK

The results of these survey measurements of the separation of trace elements from raw coal by dry magnetic separation are encouraging. Further, work carried out with the ETCi 200 Lb/Hr alpha prototype MagMill™ and MagMill™ process simulator indicates that significantly greater reductions in sulfur can be achieved efficiently with the MagMill™ than by dry magnetic separation alone as reported here.<sup>7</sup> This suggests that the potential for removal of trace elements associated with iron pyrites in coal is greater than is indicated in the exploratory measurements employing dry magnetic separation alone.

With support from the U.S. Department of Energy Solid Fuels Program, ETCi has begun an experimental program to test the feasibility of separation of trace elements, especially mercury, selenium, and arsenic, from raw coal using the alpha prototype MagMill™.

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<sup>7</sup> R. R. Oder and Y. Feng, “Dry Coal Cleaning in a MagMill,” U.S. Department of Energy Federal Energy Technology Center 1997 Coal Liquefaction and Solid Fuels Contractors Review Conference, September 2-4, 1997, Pittsburgh, PA.