

IN-MINE VARIATION AND ITS EFFECTS ON COAL GASIFICATION

Scott F. Ross and David R. Kleesattel

University of North Dakota Energy Research Center
Box 8213, University Station
Grand Forks, North Dakota 58202

Abstract

As reported earlier (1), four different lithologic layers have been identified in the Freedom Mine (Mercer County, North Dakota) which supplies the lignite for the Great Plains Gasification Associates plant in Beulah, North Dakota. The layers were identified on the basis of readily observable megascopic characteristics including luster, fracture characteristics and the presence of clay and silt zones. Lignite sampled from each of the four layers has been pyrolyzed in a bench scale reactor system designed to simulate the production of gas liquor condensate from the pyrolysis zone of an actual gasifier. The yields of water-soluble organic effluents from each of the layers were found to differ significantly, particularly the yields of phenol, cresol and catechol.

Introduction

The treatment and removal of water-soluble organic effluents from wastewater is an important issue facing coal gasification technology. The extent of treatment is governed by the reuse or environmentally acceptable disposal of the wastewater. Downstream effluent treatment is also dependent on the nature and quantity of tars pyrolysis and devolatilization reactions in the upper portion of the gasifier. It is desirable to develop a laboratory test to simulate the production of water-soluble organic effluents from a gasifier, thereby eliminating expensive pilot-plant tests. Such a test could eventually be a method of assessing the gasification potential of various coals, and the resulting data base would be helpful in designing effluent treatment systems for gasification plants.

In working towards the development of such a test, the technique was found to be sensitive to changes in coal quality which occur within the same mine. With the discovery of distinct lithologic layering within a mine which supplies coal to an actual commercial gasifier, an investigation into the effects of in-mine variation on coal gasification was initiated.

Experimental

A laboratory scale tubular reactor was constructed which allows for the pyrolysis of up to five grams of coal in a variety of gas atmospheres. A Lindbergh split-type furnace with a maximum temperature of 1100 C and a programmable heating rate of 5°C/min. to 45°C/min. allowed for reproducible heating of the samples. A liquid nitrogen cooled trap was used for the collection of water-soluble organic effluents. After completion of the experiment the trap was allowed to warm to above 0°C and the water-soluble organics analyzed by gas chromatography (2).

The coal samples used in this study were collected at the Freedom Mine (Mercer County, North Dakota). The samples were ground to -60 mesh and pyrolyzed in a nitrogen atmosphere using a heating rate of 45°C/min. and a final temperature of 850°C.

TOSCO Material Balance Assays were provided by J & A Associates, Inc., Golden, Colorado. The procedure has been described elsewhere (3).

Standard quantitative maceral analyses (4) were performed on representative samples from each of the four lithologic layers. Lignite samples were prepared for micropetrographic analysis as described in ASTM procedures (5).

Results and Discussion

During a mine study in May 1984, major lithologic units occurring as layers in the Beulah-Zap bed of the Sentinel Butte Formation (Paleocene) were observed. The seam was subdivided into four lithologic units on the basis of overall megascopic characteristics (Figure 1). The criteria for these subdivisions were:

1. appearance of the broken surfaces of the units on a large scale as they appear in the high wall;
2. luster of the coal;
3. fracture characteristics, hardness and surface appearance of the coal on a small scale (1-10 cm);
4. presence of lithologically distinct units including thin layers of fragmental coal, clay, and silt layers and concretionary zones.

There is evidence to suggest that the units are not entirely local in extent but persist widely in the Beulah-Zap bed (6).

Lignite was sampled from each of the four layers in a vertical sequence with the samples being collected within a few meters of each other. The samples were pyrolyzed as described above and the water-soluble organic effluents were analyzed. The yields of the water-soluble organics from each of these four samples and their corresponding proximate and ultimate analyses are given in Table 1. Based on the pyrolysis yield data, the top three layers appear to be quite similar. However, layer four shows considerable differences in the yields of methanol, phenol, cresols and catechol. In fact, layer four appears to be an entirely different coal. Layer four is separated from the other three layers by a locally thin, inorganic-rich zone or clay layer, suggesting that a marked difference in the depositional environment could have occurred. The proximate and ultimate analyses for the four layers are quite similar, however, and provide no explanation as to why the fourth layer should behave so differently upon pyrolysis than the other three layers. In particular, a comparison of maf ultimate data for layers 2 and 4 shows great similarity, yet pyrolysis yields of water-soluble organics are radically different. This suggests that a plant operator could not rely on routine coal analysis as the predictor of wastewater characteristics.

The data from the TOSCO Material Balance Assays are given in Table 2. The most obvious difference is the tar yields for the four layers. There is a 44% decrease in tar yield between layer 1 and layer 2. The yields of water, CO and C₁ also differ significantly between the four layers. However, unlike with the water-soluble organic effluent data, the fourth layer doesn't stand out as being different from the other three layers.

Petrographic analyses for the four layers are presented in Table 3. Unlike the proximate and ultimate analyses, which suggest little difference between the four layers, the petrographic analyses indicate that there might be considerable organic structural differences between the layers. There exists a good correlation between catechol yields upon pyrolysis and the amount of corphuminite found in each layer. A linear estimation of the data results in a correlation coefficient of 0.92.

Table 1. Pyrolysis Yields for Four Lithologic Layers in the Freedom Mine^a

Coal	(Top) Layer 1	Layer 2	Layer 3	(Bottom) Layer 4
Compound:				
Methanol	990	1010	940	1590
Acetone	1350	1320	1490	1420
Acetonitrile	240	250	260	190
2-Butanone	360	340	420	350
Propionitrile	70	130	280	190
Phenol	2110	1720	1800	3820
o-Cresol	610	520	580	980
p-Cresol	680	570	600	1190
m-Cresol	710	630	720	1420
Catechol	990	1010	1200	3150
Proximate Analysis (as rec'd; % by wt):				
Moisture	23.51	23.11	27.93	30.62
Volatile matter	29.32	33.93	34.22	36.74
Fixed carbon	31.61	36.56	32.91	27.93
Ash 15.55	6.40	4.94	4.72	
Ultimate Analysis (maf; % by wt):				
Hydrogen	4.66	5.09	4.75	5.13
Carbon	68.20	69.14	70.37	69.32
Nitrogen	1.08	1.11	1.12	1.07
Sulfur	2.75	0.66	0.68	0.84
Oxygen	23.30	23.99	23.09	23.65

^aCompound yields are reported in micrograms/g maf coal.

Table 2. TOSCO Material Balance Assay

Fischer Assay Yields	Normalized Values (Moisture Free)			
	Layer 1	Layer 2	Layer 3	Layer 4
Tar (lb/ton)	119.3	67.1	103.7	89.1
(gal/ton)	14.6	8.2	12.7	10.9
Gas (lb/ton)	311.9	333.5	329.0	306.6
(scf/ton)	3311.0	3597.7	3627.6	3402.9
Water (lb/ton)	129.9	192.3	158.7	191.2
(gal/ton)	15.6	23.1	19.0	22.9
Char (lb/ton)	1438.9	1407.1	1408.5	1413.1
H ₂ (lb/ton)	1.22	1.11	1.33	1.14
CO (lb/ton)	30.76	37.51	36.85	40.89
CO ₂ (lb/ton)	243.23	256.50	251.62	224.00
Cl (lb/ton)	17.44	19.78	22.32	20.63

Table 3. Petrographic Analyses of Freedom Mine, Four Lithologic Layers

Maceral Analysis (% Volume)	<u>Layer 1</u>	<u>Layer 2</u>	<u>Layer 3</u>	<u>Layer 4</u>
Humanite Group				
Ulminite	35.5	38.9	38.2	42.8
Humodetrinite	25.2	23.1	21.9	18.0
Gelinite	0.5	0.4	1.4	1.3
Corpohuminite	1.0	2.6	2.0	6.2
Liptinite Group				
Sporinite	0.9	2.4	1.4	3.3
Cutinite	0.7	0.5	0.5	0.5
Resinite	2.7	1.9	0.9	1.7
Suberinite	0.0	0.5	0.4	1.5
Alginite	1.2	0.4	0.9	1.2
Liptodetrinite	5.0	5.9	3.8	5.3
Fluorinite	0.0	0.4	0.0	0.0
Bituminite	0.0	0.0	0.0	2.0
Inertinite Group				
Fusinite	4.5	4.6	8.5	2.7
Semifusinite	6.8	8.1	7.2	5.3
Macrinite	0.7	0.5	0.2	0.0
Sclerotinite	0.3	0.5	0.4	0.3
Inertodetrinite	8.9	7.6	8.5	4.2
Micrinite	1.3	0.7	1.8	1.3

Conclusions

The composition of gas liquor condensate can vary greatly due to variations within an individual seam. The samples used in this study were collected within a few meters of each other but indicate significant vertical variation exists in a particular mine. The ultimate analyses of these layers are virtually identical, but the actual chemistry, as evidenced by the pyrolysis results and the TOSCO Material Balance Assays, is very different from layer to layer. These differences could result in substantial changes in wastewater composition and operability of a tar/water separator in an actual gasification plant when coal from different layers is gasified.

Petrographic analysis reflects, to an extent, the structural chemistry of the coal because the macerals generally derive from different kinds of plant constituents, and these original plant constituents in turn have different structures. Therefore, petrography should be a useful predictor of some pyrolyzate yields.

Reasonably steady operation of wastewater treatment plants and tar/water separators depend on having reasonably steady wastewater composition and tar production, or at least the ability to predict these in advance. In order to achieve this it is important to characterize the pyrolysis behavior of the coal layers to provide for blending or preferential mining and selective utilization.

Acknowledgment

This work was supported by the U.S. Department of Energy, Contract No. DE-FC21-83FE60181.

The authors would like to acknowledge John W. Diehl for doing the gas chromatographic analyses and Harold H. Schobert for his helpful comments.

Literature Cited

1. UNDERC Quarterly Technical Progress Report for the Period July-September 1984, Section 15, DOE/FE/60181-1682, November 1984.
2. Olson, E.S. and J.W. Diehl, 186th ACS National Meeting, Washington, D.C., Abstract of Papers, Anyl 63, (1983).
3. Goodfellow, Lawrence and M. T. Atwood, presented at the 7th Oil Shale Symposium, April 18-19, 1974.
4. Stach, E., Taylor, G.H., Mackowsky, M.T., Chandra, D., Teichmuller, M., and Teichmuller, R., 1982, 3rd edition, Coal Petrology, Gerbruder-Borntraegger, Berlin-Stuttgart, 535 p.
5. Annual Book of ASTM Standards, 1980, Gaseous Fuels: Coal and Coke, Part 26, D 2797-72 (1980), Preparing Coal Samples for Microscopical Analysis by Reflected Light, pp. 363-371.
6. Kleesattel, D.R., M.A. Thesis, University of North Dakota, 1985.

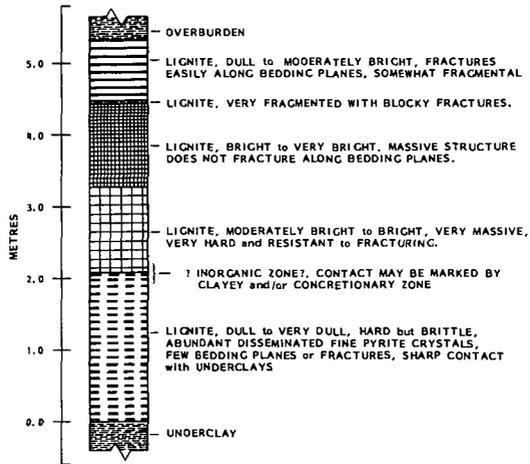


Figure 1. Lithologic units of the Beulah-Zap lignite bed as described at the Freedom Mine.