PETROGRAPHIC METHOD FOR SELECTIVE DETERMINATION OF A COMPONENT (RAW COAL) IN A MIXTURE OF PRODUCTS FROM PROCESSING OF COAL

D. M. Mason
Y. Julian

Institute of Gas Technology
3424 S. State Street
Chicago, Illinois 60616

Introduction

In the HYGAS® pilot plant gasification reactor, fine particles from each reactor stage become entrained in the gas and are carried into the exit-gas cyclone and are removed from the gas. Thus, the dust removed by the pilot plant cyclone includes particles from the steam-oxygen-gasification fluidized bed, the second-stage hydrogasification fluidized bed, the first-stage hydrogasification entrained reactor, and the slurry drier fluidized bed. (See Figure 1.) For an understanding of the process, it is desirable to estimate the amount of feed coal carried out of the reactor from the slurry drier bed. To fill this need, we have developed a petrographic method for determining the feed coal content of samples of dust collected from the cyclone. The method was developed on samples obtained when the pilot plant was operating on lignite, but it should also be applicable to samples from the processing of other ranks of coal.

If the dust collected from the cyclone were composed of material from only two sources and the two components differed in character, it would be possible to estimate their relative proportions from the elemental composition or other test property of the sample and its two sources. This situation would also apply in effect if some test property were sufficiently uniform among all sources but the one of interest. This is not the case here, as shown by analyses of typical bed samples from the different stages of the reactor (Table 1). Table 1 also shows that the cyclone dust is much finer than the samples of source materials, as might be expected. One should, therefore, analyze corresponding fines fractions of the source materials if this approach appeared fruitful. However, note that the composition of the fines elutriated from a particular bed may differ from the composition of particles of the same size range sampled from the bed. Thus, one can only surmise, from the composition data of Table 1, that the amount of feed coal in the cyclone dust is probably between zero and 25 weight percent.

Development of Method

In the customary form of petrographic quantitative analysis for the organic components (macerals) of coal, the sample is mixed with epoxy resin and pressed in a mold to obtain a cylindrical briquet. After hardening, the briquet is sectioned and polished in a manner such that the macerals can be identified under the microscope (ASTM Methods D2797 and D2799) (1). Using an eyepiece with crosshairs, successive areas of the polished surface are examined, and the macerals that appear under the crosshair intersection are counted. (A crosshair grid with multiple intersection points can also be used.) The number of counts of each component is proportional to its area in the section and its volume in the sample; with a sufficient number of counts, the percentage of counts for a component is a satisfactory measure of its volume percent in the sample. Points appearing on mineral matter and the resin matrix are ignored. The volume percent of each maceral on the mineral-matter-free basis is customarily reported, but can be converted to an ash- or mineral-matter-containing basis by calculation from ash content and estimated densities.

We concluded that application of this method to the cyclone fines would be very difficult because of the nature of some of the components other than the coal feed.

As gasification progresses the particles become increasingly porous and the proportion of mineral matter increases. Mineral matter other than iron sulfides is sometimes difficult to distinguish from the mounting medium, and clay particles especially are subject to plucking during the grinding and polishing. Instead we investigated a variant procedure in which we gravimetrically determine the weight percent of the sample in the briquet and, by a point count analysis, determine the volume percent of feed coal in the briquet, lumping all other components and mounting medium together. The densities of the feed coal and of the briquet are then needed to convert the volume percent of feed coal in the briquet to weight percent. From the weight percents of feed coal and sample in the briquet we obtain the weight percent of feed coal in the sample.

Some extra care in the mounting procedure is required to obtain briquets of known and uniform sample content. The usual technique in which the sample-resin mixture is pressed in a mold is not applicable because the clearances of the mold may allow liquid resin and some of the small particles to escape and thus change the composition of the mixture and resulting briquet. For this reason, we limit the sample content to obtain a mixture that is easily mixed and that will retain a minimum of air bubbles after hand stirring with a wooden splint. Note that the presence of bubbles is not deleterious provided they are uniformly distributed. Removing the bubbles by centrifuging, for example, is likely to cause sample concentration. Stirring with a propellor stirrer is satisfactory if done without breaking the surface of the mixture.

The density of the briquet can be determined very simply by weighing it in air and in water. Determining the appropriate density of the feed coal is not so simple. True (solid phase) density is easily determined by helium or water displacement or can be estimated from the hydrogen and ash content (3). However, some coals, especially high volatile C bituminous rank and lower, contain a substantial volume of submicroscopic pores. The density we use should include these pores but not those that are visible under the microscope, such as shrinkage cracks in lignite and the lower ranks of subbituminous coals. Particle density determined by mercury displacement is suitable if most of the particles are larger than about 100-mesh sieve size and if they do not contain an appreciable volume of microscopically observable pores or cracks. On our cyclone fines we determined true density by helium displacement on a Beckman Air Pycnometer; from this and porosity obtained from an equilibrium moisture determination, we calculated particle density. Note that use of this porosity value on our dried lignite feed is based on the conclusion, from unpublished work at IGT, that the dried lignite does not swell appreciably when immersed in water. However, we need to apply a correction to the pore volume because of the enhanced density of water (or actually of the water-coal complex) in the pores of low rank coal (4). This is the reason that density determined by water displacement on low rank coals is higher than density determined by helium displacement. Later we realized that if we had used apparent density in water, no correction would be necessary as the effect then cancels out. The respective equations for the particle density, d, of dry coal are —

$$d = [1/(d_{He} + 0.10) + M/(100 - M)]^{-1} \qquad 1)$$

and

$$d = [1/(d_w) + M/(100 - M)]^{-1} \qquad 2)$$

where d_{He} and d_w are densities in g/cm^3 determined by helium and water displacement, respectively, and M is the weight percent of equilibrium moisture. The quantity 0.10 in the first of these equations is our value for the difference between the two densities for this coal (2).

In the point count analysis of the briquet the feed coal is recognized principally by the low reflectance of its vitrinite and exinite, ranging below about 0.4%, compared with the substantially higher reflectance of vitrinite that has been heated to 800^o

to 900°F in the first stage of hydrogasification and to even higher temperatures in succeeding stages. However, some particles darken as gasification progresses in the later stages; perhaps these particles contain dispersed clay that becomes more concentrated and thus lowers the reflectance. However, these particles also become grainy, so they can still be distinguished from the feed coal rather easily. We have not been able to distinguish whether particles composed of inertinite only or mineral matter only originate from the feed coal or from one of the reaction stages. Accordingly, we count as feed coal the points falling on any maceral or on mineral matter if the particle that the point is on contains any recognizable vitrinite. Then, to correct for the presence of particles from the feed coal containing inertinite or mineral matter only, we analyze by point count the fines of the feed coal that pass a 100-mesh USS sieve to obtain the volume fraction of such particles.

The weight percent of coal in the cyclone sample is calculated according to the formula –

$$\text{Coal, wt \%} = \frac{100\ dV}{d_b(1 - I_c)W} \qquad 3)$$

where d is the particle density of the coal, d_b is the density of the sample briquet, V is the determined volume percent of coal in the sample briquet, I_c is the volume fraction of particles in the coal fines containing inertinite or mineral matter only, and W is the weight percent of sample in the sample briquet.

Apparatus and Procedures

The sample was dried enough that the retained moisture did not interfere with the curing of the epoxy resin. Weighed amounts of epoxy resin, sample, and activator were mixed to yield a briquet of accurately known and uniformly dispersed dry sample content of 30 to 40 weight percent. After curing in a mold overnight the density of the briquet was determined by weighing in air and water, and the briquet was ground and polished according to the methods of ASTM D2797 (1). Feed coal briquets were prepared in the same way.

For the point count analysis a Zeiss Universal microscope was used with a 40X Achromat objective giving a magnification of 625X with a 12.5X eyepiece. Points were counted at the corners of a Whipple disk to a total of 1000 on each of two briquets. Points were counted on the feed coal fines in the same manner except that inertinite and mineral matter were also counted.

Equilibrium moisture was determined according to a modification of ASTM D1412 (1) in which a 10 g sample was used and the temperature was maintained near room temperature by placing the equilibrium vessel in an insulated box.

Results and Conclusions

The helium density of the feed coal fines was 1.63 g/cm^3 and the equilibrium moisture content 21.1 weight percent; these give a particle density of 1.18 g/cm^3. The point count analysis of six briquets of the feed coal is shown in Table 2; two briquets each were prepared from three different coal-resin mixtures. The average content of vitrinitic particles (from the six analyses) was 91.0 volume percent. The amount of feed coal found in each briquet by the point count analysis, when calculated according to this average content of vitrinitic particles and expressed as percent of the amount determined gravimetrically in our preparation, ranged from 90% to 112% with an average of 102%.

Replicate determinations on some cyclone dusts (Table 3) indicates the repeatability that was obtained in the point count. Only one resin-sample mixture was prepared for each sample because of the limited quantity of sample available; for two of them only one briquet could be made. A new section of each briquet was exposed by regrinding and repolishing to obtain a duplicate point count analysis of each briquet.

The good average recovery on the feed coal briquets lends support to the principles of the method and indicates that systematic errors have been reduced to a satisfactory level.

With the analysis for feed coal in the cyclone dust in hand we can attempt to draw some additional conclusions about the source of the remainder of the dust. For example, if we take the analyses in Table 1 to be representative of the fines elutriated from each bed, then about 45% to 55% of the cyclone dust must come from the steam-oxygen gasifier. However, note that sampling of such streams at about 1200 psi presents severe difficulties and the analyses shown may not fully represent the composition (or size distribution) of the actual process solids.

Acknowledgments

This work was conducted as part of the HYGAS® coal gasification program jointly sponsored by the United States Energy Research and Development Administration (ERDA) and the American Gas Association. This program is under the technical direction of Mr. Stephen C. Verikios of ERDA (now DOE) and Dr. Ab Flowers of the American Gas Association.

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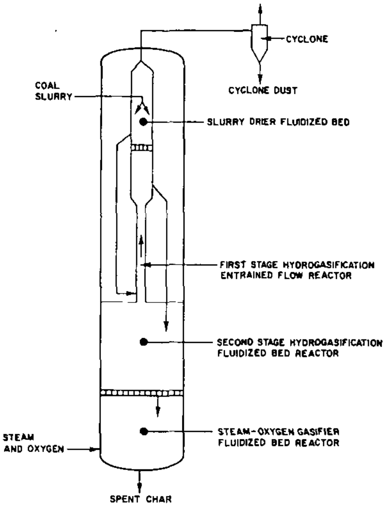

Figure 1. SCHEMATIC DIAGRAM OF HYGAS® PILOT-PLANT REACTOR SHOWING FLOW OF SOLIDS

Table 1. TYPICAL COMPOSITION OF SOLIDS IN THE HYGAS REACTOR DURING RUN 37 ON LIGNITE

	Feed Coal (Lignite)	After First Stage*†	Second Stage Bed†	Spent Char From Steam-Oxygen Gasifier†	Cyclone Dust
	wt%				
Proximate Analysis (as received)					
Moisture	17.0	3.7	2.6	7.0	2.6
Volatile Matter	35.2	19.2	8.5	9.3	15.5
Ash	9.7	22.9	28.2	46.1	36.5
Fixed Carbon	38.1	54.2	60.7	37.6	45.4
Ultimate Analysis (dry basis)					
Carbon	61.9	62.8	65.2	44.9	52.9
Hydrogen	4.31	2.70	1.56	0.94	2.00
Nitrogen	1.01	1.02	0.55	0.22	0.67
Sulfur	0.86	0.48	0.17	0.10	0.38
Ash	11.74	23.76	28.93	49.58	37.52
Oxygen (by difference)	20.18	9.24	3.59	4.26	6.53
Sieve Analysis, USS					
Retained on No.					
12	11.0	2.5	6.5	0.1	0.0
20	23.6	11.5	19.6	2.9	0.0
30	11.1	6.7	9.7	4.4	0.6
40	10.3	6.3	10.7	6.4	0.6
60	14.9	13.4	16.9	16.5	2.1
80	6.9	7.4	8.4	11.4	2.5
100	2.8	5.5	4.3	7.6	3.5
200	8.3	16.9	13.2	19.7	24.7
325	4.1	15.4	7.2	12.5	30.9
Pan	7.0	14.4	3.5	18.5	35.1

*Sampled from the spouting bed above the lift line reactor.

†Because of high-pressure sampling difficulties, these analyses may not be representative of the composition or size distribution of the process solids.

Table 2. ANALYSIS OF FEED COAL

Briquet No.	1A	1B	2A	2B	3A	3B
Density of Briquet, g/cm^3	1.216	1.210	1.198	1.198	1.209	1.209
Feed Coal in Briquet, as Prepared, wt %	33.7	33.7	28.8	28.8	26.5	26.5
Point Count Analysis						
Vitrinitic Particles, vol % of Briquet	28.5	31.9	29.9	27.2	24.6	27.1
Vitrinitic Particles, % of Whole Coal	88.8	92.1	94.0	91.0	92.1	91.2
Whole Coal, wt % of Briquet*	30.2	34.0	32.2	29.2	26.2	28.9
Whole Coal by Point Count/ Whole Coal by Gravimetric Preparation, %	90	101	112	102	99	109

*Based on average vitrinitic particle content of whole coal = 91.6 vol %.

Table 3. ANALYSIS OF CYCLONE DUSTS

	Briquet	Feed Coal Content, wt %		
Sample No.	No.	Initial	Reground	Average
1	1013	5.0	4.4	4.7
2	1015	8.2	6.3	6.6
	1016	5.7	6.0	
3	1014	5.8	4.5	5.2
4	1005	2.9	3.2	4.4
	1008	6.6	5.0	
5	1007	6.6	6.6	7.1
	1009	7.1	8.1	
6	1011	7.8	8.5	8.0
	1012	7.1	8.6	
7	1006	7.0	7.3	7.8
	1010	9.3	7.4	