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SULFUR REDUCTION OF ILLINOIS COALS

WASHABILITY TESTS

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ABSTRACT

As sulfur in the coal and oil burned in power plants is the major source of sulfur dioxide emitted to the atmosphere, the removal of sulfur from coal before it is burned would help to reduce air pollution. To determine how much the sulfur content could be reduced by preparation techniques, 40 coal samples from 35 Illinois coal mines were studied at the Illinois State Geological Survey.

Only in those Illinois coals having relatively low sulfur content, as mined, could the sulfur content be reduced to 1.5 percent or less by washing techniques. Most Illinois coals, as mined, contain 3 to 5 percent sulfur. Laboratory tests revealed this could be reduced to about 2.5 to 4 percent.

INTRODUCTION

When coal is burned under normal conditions, practically all of the sulfur in the coal is released into the atmosphere as sulfur dioxide (SO_2). Reduction of sulfur dioxide is a major objective of many current pollution control activities. One obvious way to reduce the emission of sulfur dioxide from coal-burning equipment is to reduce the sulfur content of the coal to

the practical minimum, but more information is needed to determine what that practical minimum is. The investigation described in this preliminary report and other studies are being conducted by the Illinois State Geological Survey to help provide some of the needed information. Present and projected needs for coal as fuel for electric power generation make it imperative that we learn everything possible about our coals.

Most of the coals currently mined in Illinois are cleaned before they are sold. The term "cleaned" (or "washed") indicates that the coal, as mined, is processed in a coal preparation plant to reduce the amount of ash and sulfur in the coal and increase its heating value.

The preparation process is based on the fact that mineral matter (impurities) has a higher specific gravity than the associated coal. Unfortunately, some of this mineral matter is intimately mixed with the coal and cannot be readily separated.

Washability studies are conducted in a laboratory to determine the potential reduction of ash or sulfur for a coal from any given location. Basically, this is done by placing samples of coal in solutions that have suitable specific gravity and determining the percentage of the "float" and "sink" fractions. Chemical analyses also are made on the float and sink fractions. Five solutions with varying specific gravities ranging from 1.23 to 1.60 were used to determine the relation between the chemical analyses and the percentage of sink (commonly called "reject" or "refuse"). The relation between the percentage of float (commonly called "recovery") and the chemical analyses were determined in the same manner. Laboratory float-sink data generally provide results that represent the maximum cleaning capabilities of a particular coal.

This report briefly outlines the first phase of a continuing investigation of the washability of Illinois coals and summarizes results. A more comprehensive report will be prepared and published at a later date. Meanwhile, detailed information is available at the Illinois State Geological Survey for those who need it immediately.

Acknowledgments

The U. S. Public Health Service, Department of Health, Education and Welfare, through contract No. PH 86-67-206, provided substantial support for this study. We are also indebted to the operating coal companies of Illinois for their invaluable assistance in obtaining samples.

PURPOSES OF THE INVESTIGATION

The primary aim of this investigation was to study the washability characteristics of Illinois coals, with particular emphasis on the quantity, distribution, and varieties (forms) of sulfur occurring in Illinois coals. The studies could provide a basis for determining how much pyritic sulfur could be removed from Illinois coals by conventional coal cleaning methods and also for evaluating the pyritic and total sulfur contents of coal mine refuse.

The data developed in this study supply information required for (1) determination of how much sulfur can be removed from coal from various seams, or different areas within the same seam, by conventional preparation techniques; (2) evaluation of the possibility of recovering pyrite from mine refuse to reduce its potential as a stream pollutant; and (3) evaluation of the use of Illinois coal refuse as a source of pyrite for sulfur recovery or sulfuric acid manufacture.

A secondary object of the investigation has been a comprehensive study of other chemical and physical properties of Illinois coals, including trace elements, coal petrography, palynology, and mineral matter other than sulfur compounds.

PROCEDURE

Samples

The 40 coal samples described in this report were taken from 35 Illinois coal mines located in most mining areas of the state. Coal seams sampled were the Danville (No. 7), Herrin (No. 6), Harrisburg or Springfield (No. 5), Summum (No. 4), Colchester (No. 2), De Koven, Davis, and Rock Island (No. 1). The number of samples is greater than the number of mines because several mines produced coal from more than one seam. In all cases, different seams were sampled separately.

An effort was made to obtain samples that were representative of the coal processed by the preparation plant during one full shift of operation. The ideal sample was 2000 pounds of coal made up of 20 to 30 increments taken at uniform intervals throughout a working shift. However, at many mines fewer increments were taken because sampling conditions were poor. Some samples, therefore, may not be representative of the output for the operating shift and should not be considered representative of the output of the mine for an extended period of time. The samples are, however, superior to grab samples. At several strip mines, coals from different seams were being mixed at the preparation plant, and it was not practical to obtain a single-seam sample from the tipple, making it necesary to cut them from freshly exposed faces in the pits.

In addition to the 2000-lb sample, at least three channel samples of about 50 lbs each were cut from the coal face in different areas of every mine sampled.

Laboratory Preparation

The channel samples were crushed and riffled in the laboratories to obtain adequate samples for chemical and physical analyses. Chemical analyses included proximate, heating value, total sulfur, forms of sulfur (pyritic, organic, or sulfate), and chlorine analyses. These same analyses, plus ultimate analyses and determinations of the free-swelling index, ash fusion temperature, and Gieseler plasticity were made on a composite of the three face-channel samples.

TABLE 1 - PYRITIC SULFUR[1] REDUCTION IN FLOAT COALS

Sample no.	Original sample	Float coal fraction			
		40 percent recovery		80 percent recovery	
	Pyritic sulfur (%)	Pyritic sulfur (%)	Reduction[2] in pyritic sulfur (%)	Pyritic sulfur (%)	Reduction[2] in pyritic sulfur (%)
01	2.25	0.51	77.4	1.06	52.7
02	1.94	0.70	63.9	0.82	57.6
03	0.59	0.39	34.0	0.51	14.0
04	1.70	0.72	57.6	1.15	32.1
05	0.53	0.28	47.3	0.40	24.5
06	2.45	0.44	82.0	1.12	54.1
07	2.86	1.08	62.4	1.52	46.8
08	2.25	0.60	73.3	0.89	60.4
09	2.76	0.51	81.5	1.08	61.0
10	3.35	0.42	87.6	1.25	62.6
11	1.47	0.38	74.3	1.00	31.7
12	0.68	0.25	63.2	0.68	0.0
13	2.43	0.61	75.0	1.28	47.5
14	2.28	0.58	74.6	0.87	61.8
15	2.80	0.87	69.0	1.41	49.7
16	2.59	0.59	77.4	1.11	57.1
17	2.27	0.52	77.1	1.29	43.1
18	1.20	0.26	78.2	0.58	51.6
19	2.94	0.25	91.5	1.06	64.0
20	2.99	0.82	72.7	1.82	39.2
21	2.16	0.31	85.5	0.88	59.2
22	0.80	0.29	63.2	0.40	50.0
23	2.73	0.38	86.2	0.99	63.7
24	2.82	0.32	88.7	0.61	78.4
25	1.76	0.83	52.8	1.58	10.1
26	2.87	0.56	80.4	0.88	69.3
27	2.60	0.43	83.4	1.48	43.2
28	3.30	0.88	73.2	1.65	49.9
29	3.39	1.44	57.4	2.29	32.5
30	2.91	0.39	86.6	1.27	56.3
31	2.71	0.42	84.6	0.80	70.6
32	2.55	0.49	80.8	0.93	63.6
33	2.39	0.58	75.6	0.97	59.3
34	2.22	0.40	82.0	1.19	46.6
35	2.66	0.51	80.7	0.91	65.8
36	2.89	0.34	88.3	1.12	61.1
37	4.46	0.85	80.9	1.25	72.1
38	3.34	0.64	80.8	1.60	52.0
39	2.27	0.52	77.2	1.02	55.1
40	1.15	0.49	57.0	0.74	35.4
AVERAGE	2.36	0.55	76.8	1.09	53.9

[1]Chemical data on dry basis.
[2]Percent reduction from original sample.

The 1-ton sample of raw coal was crushed to a maximum size of 1½ in. with a jaw crusher. A representative portion (about 200 lbs) of this 1½ in. x 0 coal was crushed to a top size of 3/8 in., then screened into 3/8 in. x 14 mesh, 14 mesh x 100 mesh, and 100 mesh x 0 fractions. Washability studies were made on the 1½ in. x 0, 3/8 in. x 14 mesh, and 14 mesh x 100 mesh fractions.

Gravity separations of very fine coal (less than 100 mesh) are difficult with any coal and are particularly difficult with Illinois coals, which are quite porous. The liquid used for the separation rapidly penetrates the pores and thereby exerts a major influence on the results. Although some tests were made with the 100 mesh x 0 coals, the results were not satisfactory.

Chemical Analyses

Chemical analyses, which determined percentages of ash, sulfate sulfur, pyritic sulfur, organic sulfur, and total sulfur, were made on all float fractions and on some sink fractions. Grindability and fusibility of the ash were also determined for the lightest, intermediate, and heaviest (1.60 specific gravity) float fractions of the 1½ in. x 0 coal, although the values are not presented in this report. The proximate analyses and determinations of total sulfur, forms of sulfur, heating value, and free-swelling index were made for the raw coal, as received, and on the sized fractions.

All chemical values presented in this report are on the dry basis.

RESULTS

Sulfur Removal

Three forms of sulfur—sulfate, organic, and pyritic—were determined by chemical analyses. The sulfate sulfur content is quite low, with an average of only 0.08 percent for the 40 samples tested. The organic sulfur is in complex combination with other organic matter in the coal and cannot be

TABLE 2—EFFECT OF COAL SIZE ON SULFUR REDUCTION*

Recovery (%)	Total sulfur (%)			Pyritic sulfur (%)		
	1½" x 0	3/8" x 14 mesh	14 x 100 mesh	1½" x 0	3/8" x 14 mesh	14 x 100 mesh
40	2.35	2.33	2.24	0.55	0.52	0.39
50	2.43	2.40	2.33	0.62	0.58	0.48
60	2.54	2.48	2.46	0.73	0.68	0.61
70	2.69	2.60	2.60	0.89	0.81	0.78
80	2.87	2.75	2.78	1.09	0.97	0.99

* Sulfur values are given on the dry basis and are averages of 40 samples.

TABLE 3 - CHEMICAL ANALYSES[1] OF ORIGINAL SAMPLE AND AT TWO RECOVERIES

Original sample				Float coal fraction							
				40 percent recovery				80 percent recovery			
Sample no.	Total sulfur (%)	Pyritic sulfur (%)	Ash (%)	Sample no.	Total sulfur (%)	Pyritic sulfur (%)	Ash (%)	Sample no.	Total sulfur (%)	Pyritic sulfur (%)	Ash (%)
5	1.18	0.53	13.8	5	0.79	0.28	3.2	3	0.88	0.51	7.3
22	1.37	0.80	16.1	3	0.84	0.39	4.3	5	0.94	0.40	6.0
3	1.37	0.59	19.9	12	0.85	0.25	3.8	22	1.03	0.40	6.5
12	1.43	0.68	19.9	22	0.98	0.29	4.2	12	1.41	0.78	8.7
18	1.71	1.20	23.0	18	1.21	0.26	4.6	18	1.45	0.58	8.8
40	1.74	1.15	24.6	40	1.25	0.49	4.1	40	1.52	0.74	8.1
25	2.61	1.76	19.7	4	1.44	0.72	4.2	2	1.88	0.82	5.6
4	2.79	1.70	15.1	25	1.50	0.83	3.6	4	1.94	1.15	6.8
17	3.38	2.27	17.5	2	1.69	0.70	3.0	8	2.40	0.89	7.3
2	3.58	1.94	10.5	8	2.03	0.60	4.8	25	2.48	1.58	7.4
8	3.71	2.25	15.9	17	2.08	0.52	6.9	17	2.56	1.29	9.5
11	3.94	1.47	18.5	7	2.08	1.08	4.1	31	2.57	0.80	6.8
21	4.07	2.16	21.2	30	2.15	0.39	6.5	19	2.79	1.06	11.3
14	4.13	2.28	18.3	37	2.17	0.85	4.0	37	2.88	1.25	5.4
31	4.28	2.71	16.1	10	2.25	0.42	5.5	35	2.93	0.91	8.8
30	4.36	2.91	22.5	31	2.28	0.42	3.6	7	2.95	1.52	7.5
27	4.38	2.60	13.1	6	2.30	0.44	3.0	16	2.95	1.11	9.9
9	4.48	2.76	17.1	27	2.38	0.43	5.2	14	2.96	0.87	7.9
6	4.48	2.45	15.0	21	2.39	0.31	7.1	30	2.96	1.27	11.1
20	4.49	2.99	40.9	23	2.51	0.38	6.5	21	3.03	0.88	10.0
33	4.52	2.39	19.4	11	2.51	0.38	7.9	6	3.03	1.12	5.9
16	4.58	2.59	18.5	28	2.51	0.88	8.1	10	3.04	1.25	9.5
13	4.69	2.43	20.8	19	2.53	0.25	4.1	9	3.08	1.08	8.3
7	4.76	2.86	22.2	9	2.53	0.51	4.4	28	3.15	1.65	11.6
35	4.78	2.66	21.7	14	2.60	0.58	4.2	11	3.16	1.00	11.5
19	4.78	2.94	36.1	20	2.63	0.82	4.6	24	3.16	0.61	7.7
34	4.87	2.22	14.8	16	2.67	0.59	5.5	23	3.27	0.99	10.8
39	4.88	2.27	13.9	35	2.68	0.51	4.6	27	3.32	1.48	8.0
1	4.91	2.25	16.3	38	2.78	0.64	3.2	33	3.33	0.97	9.8
23	4.98	2.73	19.6	36	2.80	0.34	5.7	26	3.36	0.88	7.5
24	5.04	2.82	14.2	33	2.97	0.58	6.3	36	3.40	1.12	8.6
28	5.08	3.30	19.1	26	3.02	0.56	4.9	15	3.49	1.41	8.8
15	5.08	2.80	15.1	24	3.03	0.32	4.3	38	3.65	1.60	5.5
10	5.18	3.35	16.4	34	3.04	0.40	5.2	39	3.66	1.02	8.7
26	5.32	2.87	14.7	15	3.07	0.87	5.3	1	3.81	1.06	8.7
29	5.33	3.39	19.7	13	3.27	0.61	5.5	13	3.91	1.28	9.2
36	5.36	2.89	14.5	1	3.35	0.51	5.1	34	3.96	1.19	7.9
32	5.38	2.55	17.0	39	3.40	0.52	5.3	20	3.99	1.82	11.9
38	5.92	3.34	12.6	29	3.46	1.44	6.5	32	4.23	0.93	8.3
37	6.19	4.46	15.0	32	3.97	0.49	5.0	29	4.26	2.29	9.8

[1]Chemical data on dry basis.

removed from the coal by physical means. The pyritic sulfur, which can be partly separated from the coal by gravity methods, is the form of sulfur of particular interest in this study.

As pyrite is about four times heavier than the associated coal, gravity methods of separation can be used. However, some of this sulfur is generally finely disseminated throughout the coal and cannot be readily separated. Table 1 shows the percentage of pyritic sulfur removed from the samples in this investigation with 40 and 80 percent recoveries (1½ in. x 0 size). Although 78.4 percent of the pyritic sulfur was removed from sample 24 with 80 percent recovery of the raw coal sample, the average removal of pyritic sulfur from all samples was only 53.9 percent. Even with 40 percent clean coal recovery, which is far less than the commercially acceptable recovery level, an average of only 76.8 percent of the pyritic sulfur was removed.

Effect of Coal Size on Sulfur Reduction

The effect of coal size upon the washability characteristics of the coal may be important. Table 2 lists the average percentages of total and pyritic sulfur (dry basis) at five recovery values and three size ranges for the 40 coals tested. The percentages of total and pyritic sulfur were usually lower in the finer coal sizes, but the differences were not great enough to consider fine grinding of these coals as an effective procedure for sulfur reduction. However, a few exceptions to this average trend indicate that fine grinding might produce a beneficial effect on some coals.

Sulfur vs. Recovery

One of the basic aims of these washability studies is to provide information that shows the potential of Illinois coals as a source of fuel with certain sulfur limits and with practical limits of recovery of the raw coal. Although the percentage of allowable sulfur may be regulated by government bodies, a practical recovery limit will vary with every mine and from day to day at any mine.

Table 3 gives the percentages of total sulfur, pyritic sulfur, and ash in the original coal samples at both 40 and 80 percent recoveries. The data are based on analyses obtained from the 1½ in. x 0 coal samples. The samples are arranged in ascending order of total sulfur percentages. The data were calculated by an IBM 360 computer, which was programmed to construct "best fitting" curves and to interpolate to obtain the sulfur and ash percentages at any desired recovery percentage.

Table 3 shows that five of the 40 coals tested could be floated with an 80 percent recovery and a maximum of 1.50 percent sulfur (dry basis). These five coals were naturally occurring low-sulfur coals that ranged from 1.18 to 1.71 percent sulfur, as sampled. Three additional coals could be reduced to a sulfur content of 1.5 percent or less with 40 percent recovery. Of all the coals tested, only 10 samples at 80 percent recovery and 19 samples at 40 percent recovery had sulfur percentages of 2.5 or less.

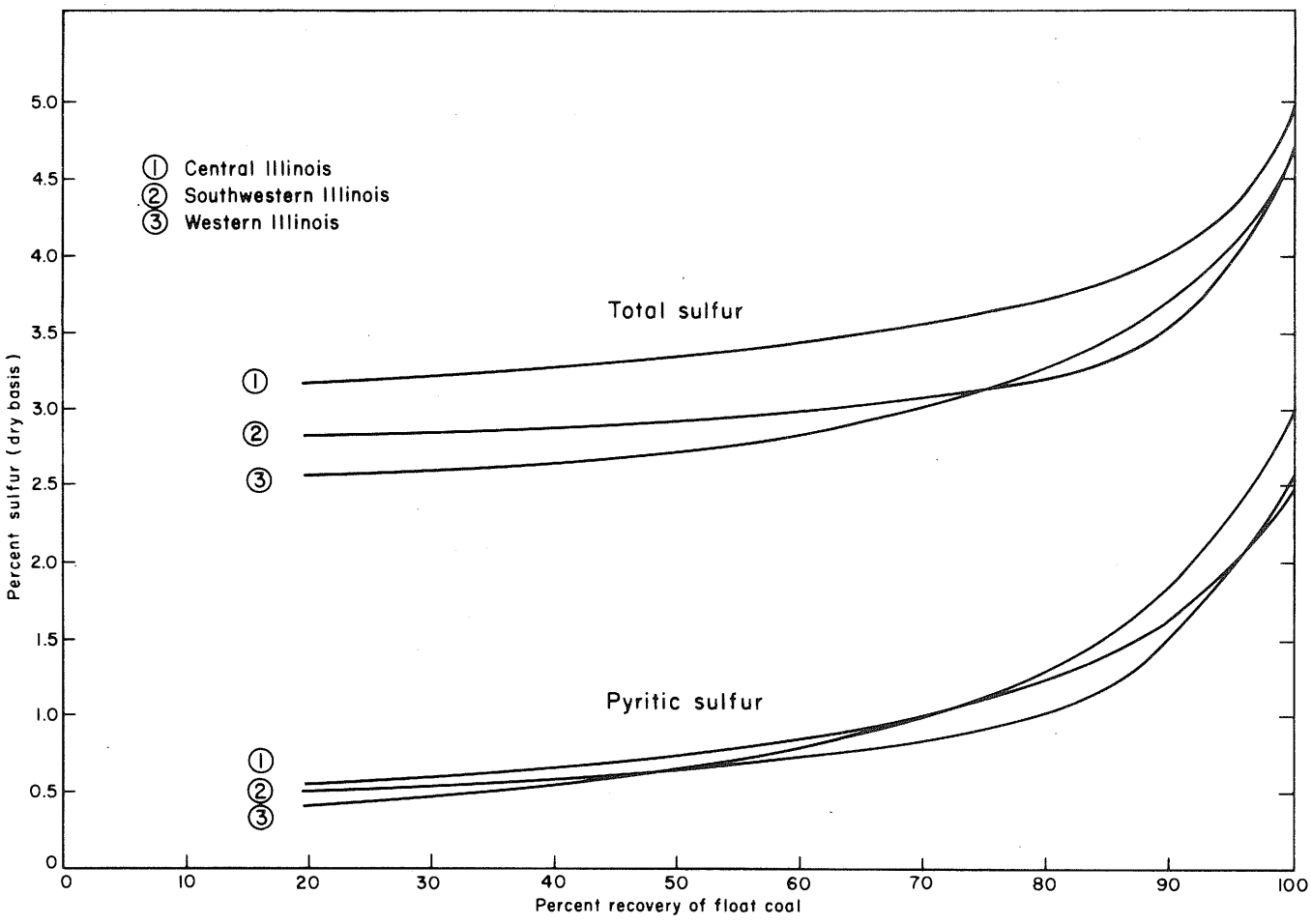

Fig. 1 - Sulfur distribution calculated for composite samples from three geological areas.

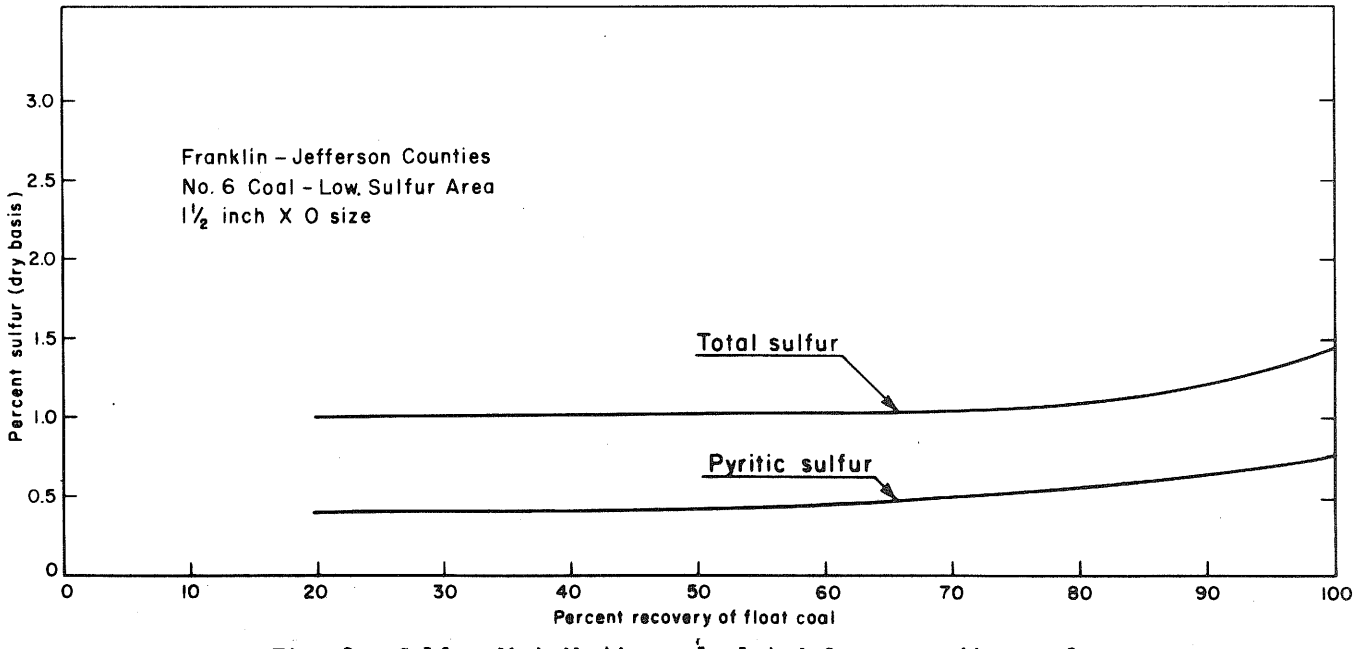

Fig. 2 - Sulfur distribution calculated for composite samples.

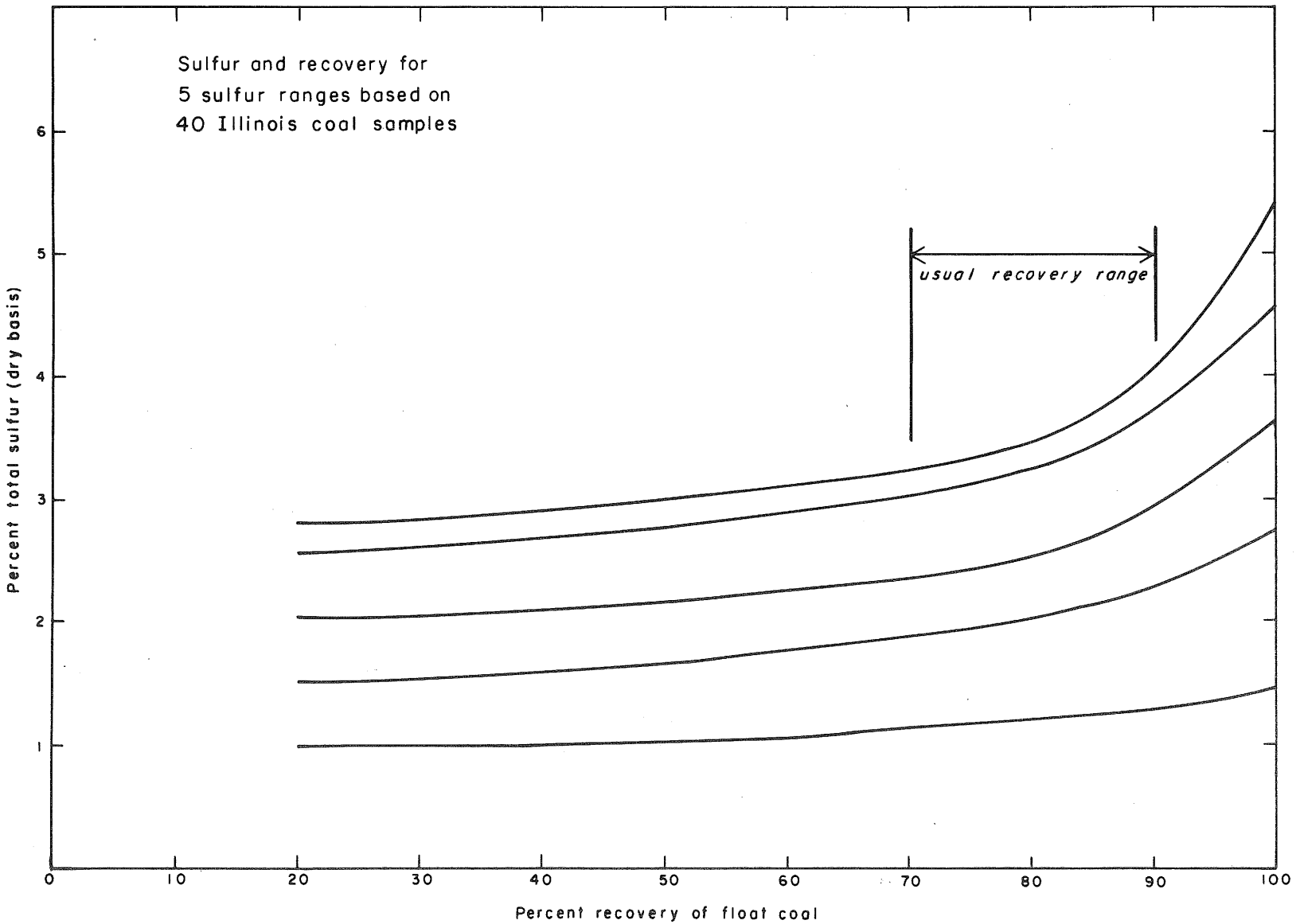

Fig. 3 - Relationship of sulfur and recovery for 40 samples.

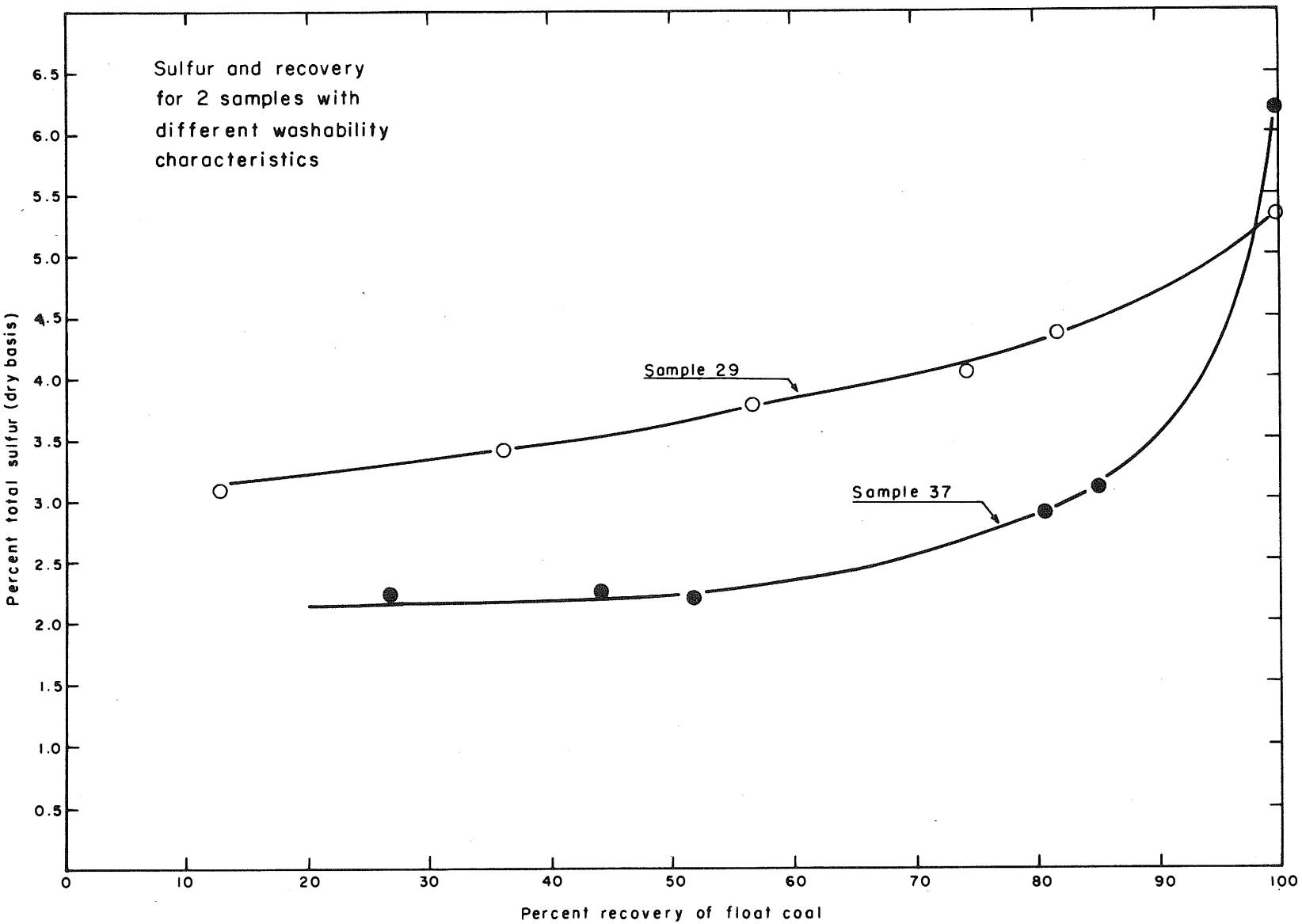

Fig. 4 - Relationship of sulfur and recovery for 2 samples.

These data should not be extended from number of samples to relative quantity of coal being mined or coal reserves available. Some of the samples are from areas with very low coal reserves, while the reverse is true of others. The annual production from the mines sampled also varied greatly. It ultimately may be practical to make quantitative estimates for total coal reserves by sulfur categories, but at present this capability is limited by lack of available samples from areas with no operating mines. Available information leads to the conclusion, however, that less than 3 percent of the total Illinois coal reserves can be cleaned to less than 1.5 percent sulfur with a normal recovery.

The upper part of figure 1 shows the average relation between the total sulfur and the float coal recovery for the coals sampled in central Illinois, southwestern Illinois, and western Illinois. The average sulfur content in the coal, as mined (100 percent recovery), was slightly less than 5 percent for all three areas. The sulfur in the coals sampled from southwestern Illinois and western Illinois was reduced to about 3 percent with a 70 percent recovery. The average sulfur in the central Illinois coals sampled was reduced to about 3.5 percent at 70 percent recovery.

The lower part of figure 1 shows the relation between the pyritic sulfur and recovery for the same coals. A remarkable similarity is shown among the coals from the three areas, with a reduction from about 2.5 percent pyritic sulfur, as mined, to about 1 percent pyritic sulfur with 70 percent recovery.

The average sulfur content for the coals sampled in the low-sulfur coal area of Franklin and Jefferson Counties is shown in figure 2. The coals, as sampled, had an average of less than 1.5 percent total sulfur. This was readily reduced to 1.25 percent with a 90 percent recovery. The pyritic sulfur varied from about 40 to 50 percent of the total sulfur.

Figure 3 summarizes the relation between the percentages of total sulfur and float coal recoveries in five sulfur ranges for the 40 Illinois coals sampled for this study. All the coals tested that contained between 1 and 2 percent sulfur, as sampled, were considered as a group, and the average values were plotted. The same was done with the 2 to 3, 3 to 4, 4 to 5, and plus 5 ranges. The pattern of reduction of sulfur is quite similar in each range.

It is important to note that figures 1, 2, and 3 represent average data from several mines. The data from individual mines sometimes varied considerably from the average. Test results from several coal samples obtained from a single mine might also show some variability.

Figure 4 illustrates the extreme variability of results between mines. The mine represented by sample 37 had more than 6 percent sulfur in the raw coal sample, yet the 80 percent float fraction had about 3 percent sulfur. The mine represented by sample 29 had less sulfur (about 5 percent) in the raw coal than sample 37, but the float coal had more than 4 percent sulfur at 80 percent recovery. The lightest 10 percent fraction of sample 29 still contained more than 3 percent sulfur.

Sulfur in the Sink Fraction

The reject material from some Illinois coal preparation plants is a possible source of recoverable sulfur. For example, the 1.60 specific gravity sink material (refuse) from one of the 40 samples tested during the present investigation contained about 26 percent sulfur. Four other coals sampled had more than 20 percent sulfur in the 1.60 sink fraction.

CONCLUSIONS

The Illinois coals sampled from active mines and tested for this study indicated that only a few could be prepared to a sulfur content of 1.5 percent or less. These samples were relatively low in sulfur when mined.

Most Illinois raw coals appear to contain total sulfur ranging from 3 to 5 percent. Our studies indicate most of these coals will retain from 2.5 to 4 percent sulfur with 80 percent recovery.

The float coal fractions (clean coal) usually had less sulfur when the coal was crushed to finer sizes. However, the differences were not great enough to make fine grinding a practical means of sulfur reduction for most of the coals tested.

The sulfur in the 1.60 specific gravity sink fractions (refuse) for the 40 samples included in this study indicated that five samples had a sulfur content of 20 to 26 percent and might be suitable for processing as a source of sulfur.

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