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A Combined Physical/Microbial Process for the Beneficiation of Coal

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ABSTRACT

A large-laboratory scale physical/microbial process was demonstrated for the removal of pyritic sulfur from coal. The process took place in an aerated-trough slurry reactor with a total slurry volume of 150 L. The reactor was divided into six sections, each of which acted as a physical separator and a bioreactor. The process objective was to physically remove the larger pyritic inclusions and to biodegrade the small inclusions (micropyrte). The process was continuously operated for 120 days, treating approximately 1 ton of Illinois #6 coal. Ninety percent pyrite removal was achieved at a 20% slurry concentration and a reactor residence time of 5 days. Additional research should be performed to find the optimum values for reactor residence time, slurry concentration, and process hydraulic residence time (or recycle ratio). Finding these optimum values will enable a process to be developed that will maximize the amount of coal that can be processed per unit reactor volume per unit time with the desired level of pyritic sulfur removal.

SUMMARY

A combined physical/microbial process for the removal of pyritic sulfur from coal was demonstrated at the large-laboratory scale. The physical/microbial process consisted of an aerated-trough slurry reactor with a total slurry volume of 150 L, divided into six sections, each of which acted as both a physical separator and a bioreactor. The process objective was physical removal of the larger pyritic inclusions, which would take many days to biodegrade, and biodegradation of the many small inclusions (micropyrite), which are difficult to separate physically. This combination should give high (up to 90%) pyrite removals without the excessively fine grinding needed for a purely physical process, because biodegradation does not require that an inclusion first be liberated from the coal, only that it be exposed to microbial attack somewhere at the coal particle surface.

The process flowsheet includes liquid recycle to provide the bacteria, acidity, and ferric ion needed to initiate the microbial activity on the feed coal, as well as biosurfactants that aid in slurring the coal. The coal is fed into Section 2, and solids settling out in Sections 2–6 are drained and recycled to Section 1. Besides acting as a second-stage physical separator (i.e., rougher section), growth of bacteria on the pyrite concentrated in Section 1 is another important source of acclimated bacteria for the process.

The process was operated continuously for 4 months, treating nearly 1 ton of an industry recognized hard-to-clean Illinois #6 coal supplied by the Monterey Coal Company and ground to 80% -100 mesh. A mixed culture consisting mainly of *Thiobacillus ferrooxidans*, but with some *Thiobacillus thiooxidans* and *Leptospirillum ferrooxidans*, was provided at startup, and a microbial consortium developed from this and the indigenous microbes on the feed coal. The concentration of free-swimming cells was on the order of 10^8 per mL, but showed considerable variation due mainly to attachment to the coal. The only nutrient added was small amounts of ammonium sulfate, the other requirements (e.g.,

phosphate) apparently being satisfied by minerals from the coal and the feedwater. The temperature was maintained at $24.5 \pm 3.1^\circ\text{C}$ and the pH kept between 1.8 and 2.2 by the addition of sulfuric acid. The process variables were the slurry concentration in Section 2 of the reactor w , the reactor residence time τ , and the process hydraulic residence time, τ_H (the ratio τ_H/τ is a measure of the liquid recycle).

Ninety percent reduction of pyritic sulfur was obtained at $\tau = 5$ days, $w = 20\%$ (wt/wt), with a recovery of 90% of the heating value in the feed coal. Of this total pyrite removal, 16% was separated physically and appeared in the solid waste drained from Section 1 and the remainder was oxidized microbially. Forty percent of the ash was also removed, although only 10% appeared in the solid waste from Section 1, the other 30% being pyrite and basic minerals that dissolve under the acidic conditions of the process. When coal loading was increased ($\tau = 3$ days and $w = 35\%$), American Society for Testing and Materials (ASTM) pyrite analysis of the product continued to show good (70%) removal, but this was not confirmed by analysis of total sulfur. This is probably caused by the partial oxidation of pyrite to elemental sulfur, and by a shortage of sulfur-oxidizing bacteria, which tend to be removed from the process attached to the product coal at this coal loading.

Because of the long time needed to reach steady state, only four combinations of the process variables were studied. Future work needed for process optimization will include several improvements. The aerated trough was originally designed purely as a bioreactor and its geometry needs to be optimized for physical separation. Losses of water, coal, and bacteria with the mist in the air leaving the process must be reduced. Finally, the washing step needed to remove sulfate from the product coal before combustion must be incorporated into the process to recover and recycle sufficient sulfur-oxidizing bacteria.

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A COMBINED PHYSICAL/MICROBIAL PROCESS FOR THE BENEFICIATION OF COAL

1. INTRODUCTION

1.1 Combining Physical and Microbial Processes

The overall objective of coal beneficiation is to produce a product low in sulfur and ash at the lowest possible cost and with minimum wastage of heating value. DOE's Coal Preparation Program has set a target of 90% removal of pyritic sulfur, but found that this can only be achieved for most coals by physical processes (i.e., flotation, agglomeration) if the coal is ground extremely fine.¹ Besides being expensive in itself, grinding coal to a top size of 10–100 μm complicates the downstream processes of coal separation and drying.

The requirement for fine grinding can be traced back to the fact that pyrite exists in coal as discrete inclusions with a wide distribution of sizes. The bulk of the pyrite in most coals is in the form of few, relatively large inclusions. However, there are also millions of very small inclusions (micropyrite) that make cleaning difficult for coals that contain them as a significant fraction of the total pyrite mass. It follows that the sensible way to clean coal is to remove the few large inclusions first, and then to achieve higher levels of depyritization by removing ever smaller inclusions. Table 1 shows the results of this hypothetical ideal procedure for a typical Ohio coal² (actually the average of 23 coals for which the inclusion size distribution has been measured and fit to the log-normal distribution). Thirty percent pyrite removal can be achieved by eliminating all inclusions larger than 83 μm , of which there are 4,900 per cm^3 of coal. This can readily be accomplished by a physical process. Grinding the coal to 160 μm provides sufficient liberation and the settling velocity of a particle this size is sufficient to provide adequate separation in a cyclone or similar process. Attempting to remove these large

inclusions by a microbial process would be counterproductive, since bacteria would need at least 6 days to dissolve an inclusion of 83 μm .

Obtaining 90% removal is considerably more difficult. All inclusions larger than 8 μm , of which there are over three million per cm^3 , must be removed. Float/sink processes are now impractical because of the low settling velocities of such small particles (the 3.67 h given as time for float/sink separation in Table 1 is the time needed for an 8- μm liberated pyrite inclusion to separate by 1 meter from a 8- μm coal particle by differential settling in a liquid). Surface-area dependent physical processes such as flotation or agglomeration could be used, although the coal must be ground to approximately 14 μm to provide sufficient liberation of such small inclusions. When dealing with these very small inclusions, a microbial process has two advantages. First, much less time is needed to dissolve small inclusions; about 1 day for an 8- μm inclusion depending on the fraction of its surface that is exposed to microbial attack. Second, a microbial process requires less coal grinding than a physical process. The reason for this is that liberation is no longer necessary; the inclusion need only be exposed to bacterial attack somewhere at the surface of the coal particle. The 30 μm given in Table 1 as the coal size for microbial access is the coal size at which sufficient inclusions are exposed, if the inclusions are assumed to be randomly distributed through the coal. In fact, when coal is ground, it tends to fracture along planes weakened by the presence of inclusions, exposing more pyrite to direct microbial attack. Therefore, 30 μm underestimates the coal size at which 90% removal is possible. Experiments on microbial depyritization have shown 90% pyrite removal from many different coals in the size range 100–200 mesh (75–150 μm).³

Table 1. Depyritization of a typical Ohio coal.

Percent pyrite removal	Largest remaining inclusion	Inclusions removed (#/cm ³)	Coal size for float/ sink removal		Coal size for microbial access		Time for float/ sink separation (h)	Time for biodegradation (h)
			D (μ m)	(d/D)	D (μ m)	(d/D)		
10	220	170	400	0.55	2700	0.08	0.005	352
30	83	4,900	160	0.52	560	0.15	0.034	133
50	42	45,000	81	0.52	230	0.18	0.133	67
70	21.0	360,000	40	0.52	99	0.21	0.533	35
90	8.0	3.1 x 10 ⁶	14	0.57	30	0.27	3.67	13
99	1.8	49 x 10 ⁶	3.9	0.46	6.3	0.29	72.5	2.9
99.9	0.80	140 x 10 ⁶	1.8	0.44	2.9	0.28	376	1.3
99.99	0.37	290 x 10 ⁶	—	—	—	—	—	—

Although the calculations that lead to the results in Table 1 are only approximate, the conclusions are considered qualitatively correct. Increasing pyrite removal means removing more and more inclusions of ever smaller sizes. It becomes increasingly difficult to do this physically without very fine grinding and/or considerable loss of heating value in the reject stream. On the other hand, a microbial process works best on the very small inclusions because less grinding is needed and their high surface/volume ratio means that small inclusions need less time for biodegradation. The work described in this report follows from these conclusions. The objective is to create a process in which the large pyrite inclusions are separated physically while the small inclusions are dissolved by microbial activity. This process is demonstrated on an Illinois #6 coal known to contain significant micropyrite, making it difficult to clean by conventional methods.

A combined microbial/physical process should also give improved removal of other ash-forming minerals in coal. The basic minerals (carbonates) will be dissolved in the acidic (pH 2) conditions of the microbial process, even if they are finely dispersed through the coal. However, a purely microbial process would not affect the insoluble, quartz-type minerals, which are relatively easy to remove physically.

Table 1 shows that both microbial and physical depyritization processes can potentially work better as the coal is ground finer. The coal top size for this work was therefore chosen on economic rather than technological grounds. The argument is that if the coal is to be pulverized to a size of -100 mesh before being blown into a boiler, then grinding it to this size is a normal cost of doing business that can be charged to the combustion step. Grinding smaller than this is not only expensive, but the cost must be charged to the coal beneficiation step with severe consequences for its economic feasibility. A coal top size of approximately -100 mesh was therefore chosen for this work, and operating conditions capable of giving 90% pyrite removal at this size were sought.

This leaves several questions including:

- Can a slurry of coal at this size be dewatered and dried?
- Can the physical/microbial process be carried out at the utility site or must post-process pulverized coal be transported to the utility site?
- Should the process be carried out only on the fines stream of existing coal preparation plants?

Some of these questions have been addressed in a previous report.³ Others remain for future work.

1.2 Background

The Earth, Environmental, and Life Sciences Group of the Idaho National Engineering Laboratory (INEL) has previously developed a purely microbial process for the removal of pyritic sulfur from coal. This process employed a mixed microbial culture based on the genus *Thiobacillus*, and an aerated-trough slurry reactor, with a long channel with a V-shaped bottom along which ran porous aeration tubes. The air bubbles kept the coal particles in suspension and provided oxygen and carbon dioxide needed for microbial metabolism. This type of bioreactor was chosen because it would be relatively inexpensive to build on a large scale.³ A large-laboratory scale (200-L) reactor was operated for several months during 1990. The results were described in previous reports^{4,5} and are summarized here as a starting point for the present work.

Ninety percent of the pyrite was removed from an Illinois #6 coal ground to 74% -100 mesh, at a coal residence time τ of 7 days and a slurry concentration of 20% (wt/wt). Only one-half of this removal was accounted for by microbial pyrite oxidation with the consequent appearance of sulfate in solution. The remainder was caused by settling of large liberated inclusions to the bottom of the bioreactor. Settled solids removed at the end of the operating period contained 61% ash and 26% sulfur. The process also produced slight reductions in ash (~10%) and heating value (~3%) in the product coal.

Liquid recycle was found to be critical to process performance. It provides the bacteria, ferric ion, and acidity needed to initiate pyrite oxidation in the feed coal and biosurfactants that facilitate coal wetting and the production of a uniform slurry. Recycling a fraction of the liquid leaving the bioreactor also reduces the volume of wastewater generated by the process. The recycle was quantified in terms of a hydraulic residence time.

$$\tau_H = \frac{\text{Water in the Process}}{\text{Flow of Water out of Process}}$$

With τ_H much greater than τ (i.e., a high recycle ratio) some unknown product apparently accumulated in the process to levels that inhibited microbial metabolism. The best overall process performance was achieved with $\tau_H = 14$ days (i.e., recycle ratio of approximately 1).

The main operating problem was found to be corrosion-erosion caused by the combined effects of mechanical abrasion, microbial activity, and the aerobic, acidic chemical environment. Corrosion of the aeration tubes was particularly serious because it led to nonuniform aeration and settling of the coal at some points in the reactor. The baffles originally installed to control axial mixing in the reactor aggravated these problems and were removed. The reactor then acted as a completely mixed tank, and the theoretical advantages⁶ of plug flow operation were not realized.

1.3 Reactor Improvements

The same large-laboratory scale plexiglass bioreactor (8 ft long x 10 in. wide with a base half-angle of 30 degrees) used in the previous experiments was employed in the work described here. It was, however extensively modified in light of the experience described in Section 1.2 and the analysis described in Section 1.1. The objective was to produce a reliable process that represented an optimum combination of physical separation of large pyritic inclusions and biodegradation of small ones. The modifications are shown in Figure 1 and described in more detail in Section 3.

The reactor was divided into six equal sections by baffles bolted firmly in place. Slurry overflow from one section to the next was through a single 1/4-in. hole, which greatly reduced axial mixing. Each section had its own aeration tube, airflow control, and flow meter. The reactor also had a conical solids collector in the center of the base of each section, and a ball valve through which the settled solids could be drained periodically.

The dry feed coal was added to Section 2, where it formed a slurry caused by the agitation provided by the air bubbles and the effect of the biosurfactants. Bacteria were able to act on the pyrite as the slurry flowed through Sections 2-6 and out onto the dewatering screen. In each section, some of the larger, denser particles tended to settle out. They were removed through the solids drain and added to Section 1; the latter served two important functions. In physical separation terms, Section 1 was a roughing section, where the rejected solids underwent a second separation step. Particles high in organic matter, that may have been collected inadvertently in the solids drained from the other sections, would stay in suspension and be recovered by flowing out with the slurry into Section 2. Only truly high density particles containing much pyrite and other minerals should have settled in Section 1. These solids were drained and constituted the solid waste from the process. In microbiological terms, Section 1 can be thought of as a continuous inoculum generator. The solids in Section 1, having undergone one physical separation, were enriched in pyrite and the degradation of this pyrite was accompanied by microbial growth. These microbes were added to those flowing into Section 1 with the liquid recycle stream, and they flowed out into Section 2 to inoculate the feed coal.

There are two areas of uncertainty in this process operation that would benefit from further research. The first concerns the role of what may be loosely called flotation effects in the physical separations. No frothers or other flotation chemicals were added (they are expensive and inhibit microbial activity) and no froth forms on the slurry surface. Nevertheless, the organic fraction of the coal is more hydrophobic than the pyrite and other

Figure 1. The physical/microbial process flowsheet.

mineral matter, and there is some evidence that the difference is increased by the presence of *Thiobacillus* bacteria or perhaps the biosurfactants they produce.⁷ It follows that organic coal particles are more likely to attach to the bubbles and be transported up to the surface of the slurry, from where they flow into the next section. The relative importance of these hydrophobic effects and density effects in keeping the high mineral particles down at the bottom of the reactor is unknown. It may be possible to greatly improve the physical separation by simply changing the shape of the section (e.g., increasing the depth).

The second uncertainty concerns the importance of the continuous inoculation function of Section 1. The cell concentration in the reactor is fixed by the balance between the number of cells that grow on the available pyrite, and the numbers that leave the process either in the liquid waste or adsorbed on the product coal. The growth yields for the autotrophic bacteria *Thiobacillus* and *Leptospirillum* on pyrite are quite low and their affinity for attaching to coal surfaces is quite high, so this balance may not produce a sufficiently high cell concentration to maximize the rate of pyrite biodegradation. Liquid recycle and the microbial growth in Section 1 all help this balance, but it remains a concern when dealing with low-pyrite coal. This question is difficult to resolve experimentally. Even if the total attached biomass on the product coal could be measured reliably (this remains a topic for research), there would still be a concern that the microbial culture was becoming deficient in specific critical classes of bacteria (e.g., those like *Thiobacillus thiooxidans* that are responsible for sulfur oxidation).

1.4 Comparison with the European Process

The European Economic Community (EEC) is currently building a 1 ton/d pilot plant for the

purely microbial depyritization of coal in Sardinia, Italy. Details of this pilot plant are now available,⁸ and can be compared with the process proposed here.

The EEC process also consists of six equal tanks in series. However, there is no liquid recycle and thus no input of biomass to the first reactor except what is attached to the feed coal. This limits the flow through the process because if the dilution rate (1/residence time) in the first tank exceeds the biomass specific growth rate then all the microbes will be washed out. The absence of recycle also wastes biomass unnecessarily from the process, requires a separate mixing tank to produce a uniform slurry, and produces a large volume of dilute wastewater for treatment. A process with recycle generates a smaller volume of more concentrated waste that is easier to treat.

In the INEL proposed process, the large, dense mineral particles are deliberately allowed to settle out. This reduces the energy required for mixing to keep the slurry in suspension. The EEC process recently switched from airlift, Pachuca tank reactors to mechanically agitated reactors in an attempt to reduce the energy cost of keeping the dense particles in suspension for the long times needed by a purely microbial process. Based on our experience, corrosion of the agitators is likely to be severe.

The coal separation and washing steps in the EEC process use gravity settlers for slurry dewatering. The process we have proposed³ uses pressure filters. The relative merits of these two methods are unknown, but it is certain that efficient washing of the coal to remove the sulfate produced, and to recover the bacteria, will be critical. The amount of water needed for washing must be kept to a minimum, again in order to reduce the volume of wastewater that must be treated.

2. GOVERNING EQUATIONS AND PERFORMANCE CRITERIA

2.1 Mass Balances and Measures of Performance

The process shown in Figure 1 has two inlets, the feed coal (subscript f) and the acid/nutrient stream from the reservoir (subscript r), and three outlets, the wastewater (subscript w), the product coal (subscript p) and the solids drained from Section I (subscript l). Each of these flows contains water (flow = F kg/d), coal (flow = G kg dry coal/d), and various forms of sulfur and minerals. The steady-state water balance over the process is:

$$F_f + F_r = F_w + F_p + F_l + E \quad (1)$$

The actual values of the terms in this equation are given in Appendix B1. On a typical day, over 10 kg of water (F_r) and 4 kg of coal flowed through the process. The waste solids from Section I were screened, the liquid being returned to the reactor. The water remaining in solids was of order 0.1 kg/day and can be ignored. All the other liquid flows are measured and Equation (1) is used to calculate the water loss rate E . This loss was caused by a combination of evaporation and a mist generated by the vigorous aeration of the slurry that passed through the demisting filters on the air effluent from the reactor. Only two flows in the process are directly controlled. The recycle pump controls the flow into the reactor F_o , and thus the reactor residence time ($\tau = W/F_o$), where W = mass of water in the reactor. The waste pump controls the wastewater flow, F_w , which fixes the process hydraulic residence time, defined as the mass of water in the reactor divided by the rate at which the water flows out of the process.

$$\tau_H = \frac{W}{F_p + F_l + F_w + E} = \tau \frac{F_o}{F_r + F_f} \quad (2)$$

Note that the moisture in the product coal, F_p , is fixed by the efficiency of the dewatering process, which means that τ_H has a finite maximum value when $F_w = 0$. The process was actually operated at this no wastewater limit for much of the time. An alternative to τ_H would be to think in terms of a

recycle ratio defined by $F_o = F_r(1 + R)$. It is clear from Equation (2) that if the moisture in the feed coal, F_f is negligible, then $\tau_H = \tau(1 + R)$. If there is no liquid recycle then $F_o = F_r$ and $\tau = \tau_H$.

The steady-state sulfur mass balance for the process can be written:

$$\left(\frac{FS}{3} + GX\right)_f + \left(\frac{FS}{3}\right)_r = \left(\frac{FS}{3} + GX\right)_p + \left(\frac{FS}{3} + GX\right)_l = \left(\frac{FS}{3} + GX\right)_p + \left(\frac{FS}{3}\right)_l \quad (3)$$

S is the mass fraction of sulfate in the water, D_l is the mass flow of (dry) solid waste, X is the mass fraction of pyritic sulfur in the coal (dry basis) and the 3 is a conversion factor from sulfur (atomic weight = 32) to sulfate (molecular weight = 96). The contribution of pyritic sulfur in the fines that appear in the wastewater, $(GX)_w$, was found to be negligible. E_m is the rate of water loss from the reactor caused by misting, and S_m is the sulfate concentration in the mist. This term was found to give a surprisingly large contribution to the overall sulfur balance. The $(FS/3)_r$ term results from the addition of sulfuric acid and ammonium sulfate to the feed reservoir for the purposes of pH control and nutrient addition. Sulfate is used here because other anions added to the process may either inhibit microbial activity (Cl^- , citrate), leave undesirable elements in the product coal (Cl^- , NO_3^-), or cause the formation of undesirable precipitates (PO_4^{3-}).

For the streams labelled f, p, and l, it is important to realize that S is not an actual sulfate concentration, but an operationally defined parameter. Besides dissolved sulfate, a slurry sample may contain hydroxysulfate precipitates in the liquid or attached to the coal, and sulfate sulfur from the coal itself. The convention is adopted that all of these forms of sulfate are actually in the liquid. Thus, S_f is the concentration that would result if all the sulfate sulfur in the feed coal were actually dissolved in the coal moisture. To measure S in a slurry sample, all the forms of sulfate are first dissolved in hydrochloric acid, and the coal is filtered out and

dried to determine its moisture content. The mass of sulfate in the filtrate is then measured, and S determined by dividing this mass by the liquid in the original sample. Although this definition of S is convenient for microbial desulfurization processes, it does require some care. For example, it is reasonable to suppose that dissolved sulfate concentration in the water leaving the reactor is the same as that in the wastewater and in the interstitial water in the product coal on the dewatering screen. However, it does not follow that $S_6 = S_w = S_p$, because of the effect of the other forms of sulfate. S_6 is the sulfate concentration in the water leaving the reactor (Section 6).

Two different measures are used to define the performance of the process in removing sulfur. The product quality, y , is defined by the fractional reduction of pyritic sulfur:

$$y = 1 - \frac{X_p}{X_f} \quad (4)$$

The fraction of the pyritic sulfur that is oxidized to sulfate by the microorganisms is defined by the fractional pyrite oxidation, r_o , which is the rate of appearance of sulfate sulfur divided by the rate that pyritic sulfur is fed into the process.

$$r_o = \frac{F_p S_p + F_w S_w + F_m S_m + F_1 S_1 - F_p S_f - F_p S_t}{3 G_p X_f} = \frac{G_p X_p + D_1 X_1}{G_p X_f} \quad (5)$$

The difference between y and r_o is a measure of the physical removal of pyrite in the process, which is the fraction of the pyrite in the feed coal that appears in the solid waste. Note however, that if the $D_1 X_1$ term was zero (i.e., no physical separation), r_o would be slightly larger than y because $G_p < G_f$. Some of the effects that result in this incomplete recovery of the coal mass (e.g., dissolution of pyrite and other minerals) are beneficial, while others (e.g., loss of fines to the wastewater and oxidation of the coal organic matter) are not. The best measure of this aspect of process performance is therefore the heating value recovery

$$HVR = \frac{(G\Delta H)_p}{(G\Delta H)_f} \quad (6)$$

Where ΔH is the heating value of coal.

2.2 Ash Balances and the Solids Drain Strategy

Operation of a complex process like that shown in Figure 1 raises a number of related questions. What is meant by coal residence time or slurry concentration and how is the mass of solids to be drained from each section determined? We could, for example, choose to remove an amount equal to 1/20th of the coal feed from each of Sections 2 through 6 and add it to Section 1, and then remove 1/5th of this (i.e., $\frac{5}{20} \times \frac{1}{5} = \frac{1}{20}$ of the feed) as solid waste. This would create a uniform decrease in the slurry concentration in Sections 2 through 6, and a low slurry concentration in Section 1 (this low concentration may be desirable to facilitate oxygen transfer into a slurry that contains high levels of pyrite).⁹

The above procedure would be arbitrary because it takes no account of the ash concentration in the particular coal at any point in the process. A more sophisticated operating strategy must first recognize that the ash-forming minerals in coal can actually be divided into three categories according to their behavior in the process. Basic minerals such as carbonates will dissolve in the acidic (pH 2) conditions in the reactor. Some of the insoluble minerals are finely dispersed throughout the coal and have not been liberated by grinding to the coal top size used in the process (this is the ash in the float fraction in a float/sink test minus the basic minerals). Physical separation can affect neither of these categories. The physical part of the process is designed only to remove the third category, insoluble minerals (including pyrite) that have been sufficiently liberated by grinding. For the purposes of this section, ash refers only to these insoluble, liberated minerals.

The analysis is based on the concept of a separation factor defined by:

$$\alpha = \frac{\text{ash content of coal drained from a section}}{\text{ash content of coal overflowing from a section}}$$

If this is assumed to be a constant, mass balances for total solids and ash can be written for each section and the entire reactor. Back mixing between sections is ignored, as is the slight reduction of total solids mass caused by the dissolution of pyrite and soluble minerals.

For $i = 3$ through 6, where i is the reactor section

$$\begin{aligned} G_{i-1} &= G_i + D_i \\ G_{i-1}A_{i-1} &= G_iA_i + D_i\alpha A_i \end{aligned} \quad (7)$$

A_i = ash in coal in slurry leaving section i

G_i = coal flow rate in slurry leaving section i

D_i = coal flow rate in drain from section i .

For Section 2:

$$\begin{aligned} G_f + G_1 &= G_2 + D_2 \\ G_fA_f + G_1A_1 &= G_2A_2 + D_2\alpha A_2 \end{aligned} \quad (8)$$

For the entire reactor:

$$\begin{aligned} G_f &= G_6 + D_1 \\ G_fA_f &= G_6A_6 + D_1\alpha A_1 \end{aligned} \quad (9)$$

To complete the analysis a strategy must be specified for defining how much solids should be removed from each section. It is reasonable to make this proportional to the amount of ash entering the section. Since the amount of ash entering any section i , is equal to the amount leaving ($G_iA_i + D_i\alpha A_i$), this can be written:

$$D_i = \beta A_i(G_i + \alpha D_i) \text{ for } 1 \leq i \leq 6 \quad (10)$$

where β is a process variable set by the operator and is defined as the solids drained from a section divided by ash being fed into a section. Given

perfect ash liberation and separation ($\alpha = \infty$), setting $\beta = 1$ would make all the ash in the feed appear in the drain from Section 2 and hence in the solid waste from Section 1. D_3 through D_6 would be zero because no ash would ever reach these sections. In the real world of imperfect separations these drains are needed. Setting $\beta > 1$ tends to give improved ash rejection at the expense of decreased heating value recovery.

Equations (7) to (10) constitute 18 equations that can be solved (see Appendix A) for the 18 unknowns A_i , D_i , and G_i ($i = 1$ through 6). This leaves only the problem of specifying the relationship between the coal feed rate, G_f , the slurry volume in the reactor, V , and the liquid flow rate into the reactor, F_o . Note that F_o is assumed constant through the reactor, which implies that the effects that increase the liquid mass flow rate (addition of moisture with the coal and dissolution of coal minerals) are balanced by those (evaporation and misting) that tend to reduce it. Two process variables must be specified. The slurry concentration is specified in Section 2 because this is where it is highest. As a mass fraction it is:

$$w = \frac{G_2}{G_2 + F_o} \quad (11)$$

The reactor residence time (τ) was defined in the previous section as W/F_o where W = mass of liquid in the reactor. Since the slurry concentration is different in each section this must be related to the total slurry volume, V , by:

$$W = F_o\tau = \frac{\rho_l V}{6} \sum_{i=1}^6 \frac{1}{1 + \frac{\rho_l G_i}{\rho_c F_o}} \quad (12)$$

The results of these calculations serve two purposes: (1) they provide a guide to process operation (i.e., how much solids should be drained from each section?; what are the coal and liquid feed rates needed to give a certain residence time and slurry concentration?) and (2) they also provide a baseline against which the performance of the actual process can be judged (i.e., what should be the ash content of the product coal?; what is the variation of the coal slurry concentration between sections?).

Figures 2 through 6 show the results as functions of the separation factor for conditions that approximate one set of experimental conditions. The parameters are $\tau = 5$ days, $w = 20\%$, $A_f = 5\%$ (the feed coal was 9.55% total ash, but 2.5% was soluble basic minerals and an estimated 2% was finely dispersed) $\beta = 1$, $\rho_c = 1.35$ kg/L, and $V = 150$ L. Figure 2 shows the feed rates of coal (G_f) and water (F_o) needed to achieve the specified residence time (τ) and slurry concentration in reactor Section 2 (w). As the separation factor increases the ratio G_f/F_o actually increases slightly from 0.214 with no separation ($\alpha = 1$) to 0.237 at $\alpha = 10$. Clearly as the physical separation improves, we can make better use of the available reactor volume.

Figure 3 shows the mass of solids that must be removed from each section. The amount removed from Section 2 is considerably higher than that from Section 6, because the feed to this section contains far more ash that is likely to settle out.

D_2 initially increases with the separation factor, because Section 1 receives more ash and some of it is washed through to Section 2, increasing the ash loading on this section. However, for $\alpha > 4$, the improved separation in Section 1 reduces this effect, and D decreases with increasing α for Sections 2 through 6. The solids drained from Section 1 increase slightly with α , in line with the increase in the coal feed rate (see Figure 2).

The quality of the coal drained from each section is shown in Figure 4, which shows the great advantage of the two-stage physical separation. Even if a very high separation factor ($\alpha = 10$) could be achieved, the solids drained from Sections 3 through 6 would still contain only 15–25% ash. Discarding them would represent a considerable waste of heating value. However, after the second separation step in Section 1, the drained solids would (with $\alpha = 10$) contain over 70% ash.

Figure 2. Solids flow model: coal and liquid feed rates. Variables were fixed at $W = 20\%$, $A_f = 5\%$, and $\tau = 5$ d.

Figure 3. Solids flow model: solids drain rates. Variables were fixed at $W = 20\%$, $A_f = 5\%$, and $\tau = 5d$.

Figure 4. Solids flow model: ash in drained solids. Variables were fixed at $W = 20\%$, $A_f = 5\%$, and $\tau = 5d$.

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Figure 5. Solids flow model: solids concentration in reactor. Variables were fixed at $W = 20\%$, $A_f = 5\%$, and $\tau = 5d$.

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Figure 6. Solids flow model: fractional ash removal and Btu loss. Variables were fixed at $W = 20\%$, $A_f = 5\%$, and $\tau = 5d$.

Figure 5 shows how the movement of solids between sections controls the slurry concentration (G_i/F_i) in each section. The concentration in Section 2 is fixed at 0.25 kg/kg, corresponding to a slurry mass fraction [see Equation (11)] of $w = 0.2$. There is a steady decline in concentration through Sections 2-6, although the decline is less at higher separation factors because fewer solids are drained (see Figure 3). It can be seen that the proposed operating strategy produces a low slurry concentration in Section 1. This may be beneficial because the high pyrite concentration in this section produces a higher demand for oxygen by the microorganisms, and oxygen transfer rates are lower in dense slurries.⁹ On the other hand a dilute slurry does represent wasted bioreactor volume. An improved operating strategy would be to remove more solids from Sections 2-6 (e.g., set $\beta = 2$ for these sections) while keeping the amount drained from Section 1 the same to avoid

excessive loss of heating value in the solid waste. This was, within certain practical constraints, the strategy followed during the experiments.

Figure 6 summarizes the expected performance of the physical separation process as a function of the separation factor. With no separation ($\alpha = 1$), removing 5% of the feed coal as solid waste simply removes 5% of the ash and 5% of the heating value. Increasing α to 10 gives over 70% ash removal with only 1.5% loss of heating value.

The actual value of separation factor that can be achieved in the reactor was measured before startup (Section 3.3) and later refined based on process operation (Section 4.3). Combining this measurement with Figures 2-6 (or their equivalent for other values of τ and w) allows all the solid and liquid flows around the reactor to be fixed.

3. EQUIPMENT AND PROCEDURES

The equipment and procedures used for these experiments were similar to those described previously.⁴ This section discusses the improvements made for the present set of experiments and the reasons for them.

3.1 Liquid Flows and Axial Mixing

The same plexiglass laboratory reactor, 8 ft long x 10 in. wide with a 30-degree half-angle at the base was used. The slurry level was fixed by outlet tubes, 3/8 in. ID x 4 in. long. Three tubes, corresponding to slurry volumes of 100, 150, and 200 L, were available but only the 150-L level was used.

Previous experience had shown that trying to operate a continuous, small-scale slurry reactor at conditions approximating plug flow produced a number of scale-down problems involving plugged lines and settling of coal in the reactor.⁴ The reactor had therefore been operated essentially as a continuous stirred tank and the potential advantages of plug flow coupled with cell recycle had been lost. Several improvements were made to rectify this situation.

During previous experiments the process hydraulic residence time was fixed by removing a certain volume of liquid from the outlet reservoir (see Figure 1) each day, and replacing it with fresh water containing the desired nutrients. The resulting diurnal variation in the composition of the feed to the reactor was effectively damped out by the mixing in the reactor. This would not be acceptable in a plug-flow reactor. The wastewater pump shown in Figure 1 was therefore added to pump waste continuously from the system to a reservoir that sat on a weighing scale so that the wastewater flow could be monitored. The wastewater was removed from the bottom of the outlet reservoir to maximize the removal of fines and precipitates from the system. However, the concentration of solids in the wastewater never exceeded 2% except during process upsets caused by failure of the slurry dewatering screen (a

41- μ m nylon mesh that was replaced monthly). As the wastewater flow rate was decreased, the hydraulic residence time of the process was increased [Equation (2)] up to some upper limit corresponding to zero wastewater flow (i.e., the only significant liquid effluents from the process are the water in the product coal cake and evaporation and mist losses). The process was usually operated at this limit.

The liquid level in the outlet reservoir was maintained by a float valve. When the level dropped, water containing nutrients flowed from the feed reservoir into a side chamber connected to the outlet reservoir by tubes. This chamber acted as a mixer where fresh feed was mixed with an appropriate amount of recycled liquid from the outlet reservoir before being pumped out to the reactor by the recycle pump. This arrangement of float valve and side chamber was necessary to automatically make up water losses from the process, while not allowing short-circuiting of the fresh feed directly to the wastewater. The feed reservoir also sat on a weighing scale so that the flow could be monitored. The feed consisted of dechlorinated tap water containing 0.49 g $(\text{NH}_4)_2\text{SO}_4$ per kg of coal fed and enough 36 N H_2SO_4 to keep the pH in Section 2 below 2.2. The amount of ammonium sulfate was based on a cell yield of 0.04 g cells/g pyrite¹⁰ and this coal having 1% pyritic sulfur. The amount of sulfuric acid varied between 5.5 and 7.5 mL per kg of coal fed.

Axial mixing in the reactor was reduced by the addition of solid baffles, dividing it into six equal sections, each with its own aeration system. Flow between sections was through single 1/4-in. holes, countersunk to a sharp edge to prevent coal from settling and plugging them. Each baffle had three holes, corresponding to the slurry levels of 100, 150, and 200 L, but the 100-L holes were plugged and the 200-L holes were above slurry level. This arrangement maximizes physical separation, because only solids near the slurry surface, which should be enriched in the light, hydrophobic organic fraction of the coal, can flow into the next section.

Tracer studies were conducted to assess the success of these measures to control axial mixing. The system was first filled with tap water. A concentrated salt solution was added to Section 1 (inlet end) and the electrical conductivity measured in Section 6 (outlet end), while water was pumped from the outlet reservoir into the reactor at a rate $F_0 = V/\tau$, where V = liquid volume in reactor and τ = nominal residence time.

With such long residence times and the high aeration rates needed to ensure complete mixing in each section [0.5 standard cubic feet per minute (SCFM) to each section], the loss of water caused by evaporation and misting complicated these experiments and the analysis of the data. The system was run basically as in Figure 1 with no wastewater flow, and the recycle pump feeding the reactor from the outlet reservoir. Inflow of tap water from the feed reservoir via the float valve maintained the level in the outlet reservoir and compensated for evaporation. With this arrangement, if the reactor is operating as a series of n stirred tanks, the entire system can be considered as a series of $(n + 1)$ tanks with continuous recycle of liquid from $(n + 1)$ (the outlet reservoir) back to 1. The theoretical solution for the concentration in the n th tank of such a series is available for comparison with the data, but care is needed in defining the residence time for each tank. The volume of each of the n theoretical tanks is fixed at V/n , but the flow into each is progressively decreasing because of the evaporative losses. If the total loss rate is E , the average liquid flow rate is $(F_0 - E/2)$ and the residence time of a theoretical tank can best be defined as:

$$\tau_n = \frac{V}{n \left(F_0 - \frac{E}{2} \right)} = \frac{\tau}{n \left(1 - \frac{E}{2F_0} \right)}$$

This procedure contains another inexactitude. Without knowing E and n in advance, it is impossible to give the external tank, (i.e., outlet reservoir), a residence time equal to τ_n . However, this has no effect on the results up to a time of about 1.4τ . The exact configuration of the external tank only influences the results after about 2τ , when an element of the tracer has a chance to flow com-

pletely around the system and trigger the conductivity detector for a second time. Note that the evaporation rate can be measured during the experiment by the rate at which the level drops in the feed reservoir.

The results are shown in Figure 7 as conductivity in Section 6, normalized by the conductivity corresponding to (added tracer)/(total water volume in system) versus time/ $n\tau_n$. [Note that $n\tau_n = \tau/(1 - E/2F_0)$ = residence time corrected for water loss]. Also shown are the theoretical results for $n = 4, 5$, and 6 . In comparing the data with the theory, it must be remembered that there is some uncertainty on the y-axis because of unresolved questions about the nature of the water loss. If the loss is caused by misting, then salt is being lost as well and the average salt concentration in the system decreases with time. If water loss is caused by evaporation, then no salt is lost and average tracer concentration would remain constant. With this effect in mind, the reactor appears to be operating functionally as a series of five stirred tanks. While this is still far from the ideal of plug flow, it represents a considerable improvement over the previous experiments.

The fact that a reactor divided into six sections behaves like five tanks in series, indicates that at these very long residence times, backmixing between sections through the small (1/4-in.) holes in the baffles is significant. This backmixing can be avoided in the future by not putting all of these holes (and the reactor outlet tube) on the same level. Arranging the holes to give a slightly lower level in each succeeding section would force water in section i to flow uphill to get back into section $(i-1)$.

3.2 Aeration and the Solids Drain

Each section of the reactor had its own aeration tube, pressure gauge, flow control valve, and air-flow meter feeding an aeration tube that ran along the bottom of the trough for the entire length the section. The aeration tubes must give good bubble dispersion, not leach anything inhibitory

Figure 7. Results of axial mixing study.

to the bacteria, and not plug, abrade, or corrode in the low pH conditions surrounded by liberated quartz and pyrite particles. They must also have a reasonably low pressure drop to minimize air compression costs in a commercial-scale operation. Two basic types of aeration tubes were evaluated.

The first were hollow, rigid tubes, 5/8 in. or 3/4 in. OD, to fit into the base of the trough, and of various plastics (butyrate, polycarbonate, polyethylene, and acrylic). The ends of the tubes were plugged and small holes were drilled in two rows 60 degrees apart so that the bubbles would emerge parallel to the trough walls. (Bubbles impinging on the trough walls have been found to cause rapid corrosion of the plexiglass). The holes were uniformly spaced except over the central 3 in. of tube, which had no holes. Hole sizes were specified in terms of the drill bit used to make them. The 81 bit (0.010-in.) holes were found to require excessive pressure drop and plug quickly, so most investigations used 73 bit (0.024-in.) or 66 bit (0.033-in.) holes.

The second type used two lengths of silicone rubber tubing, 3/32-in. wall thickness, stretched over hollow rigid plastic formers of 1/2-in. OD. The formers had their ends plugged and slots cut along the side to permit airflow. Holes were punched in the silicone rubber tubing in the same configuration described above using an 18-gauge needle.

Figure 8 shows a cross section through the solids drain in the base of the reactor at the center of each section. These drains consist of a cone with a 15-degree half-angle tapering down to a 1/2-in. nipple pipe thread (NPT) line containing a PVC ball valve to remove the accumulated solids. There was no aeration directly above these drains, so Figure 8 shows only the rigid former of the aeration tube, the tube bringing the air down into it, and a small tube that directed some air down into the cone in an attempt to keep the accumulated solids there loose and free-flowing. The latter was not successful, and the accumulated solids would only flow out some time after the valve

was opened, unless they were dislodged in some manner. This aspect of solids handling needs improving in future designs.

One objective in designing the reactor sections was to maximize the physical separation of mineral matter. A pattern of mixing and recirculation must be created that keeps the organic coal particles in suspension while the denser and more hydrophilic particles settle out into the solids drain. Each section must contain regions where water and light hydrophobic particles are entrained with the rising swarm of bubbles; the upflow here being balanced by downward flow of liquid and solids in other regions (mainly near the walls). Having no aeration immediately above the solids drain was a deliberate decision; a down-flow region here should deposit denser particles in and around the solids drain while the lighter particles follow the liquid streamlines and get reentrained in the surrounding upflow regions.

In some early tests, a shallow bed of coarse particles (20 x 40-mesh sand or 0.5-mm glass beads) was added to the bottom of the trough, filling the conical drain and just covering the aeration tube. The aeration kept this bed ebullient, and it was hoped that the settled pyrite and mineral particles would mix with it while the coal particles were kept away from the aeration tube. Solids removed from the drain could be wet sieved to separate the sand for return to the trough. Although this idea worked and high-ash solids could be separated from the sand, long-term operation showed no improvement in tubing life and had a tendency for the sand to cement together, plugging the drain. The idea was therefore abandoned.

3.2.1 Physical Ash Separation

Complete optimization of the design for physical separation was not possible within the time and funding constraints of the project. Only short experiments could be done to investigate the variables most likely to affect the physical separation of ash and pyrite; the airflow rate and the number and size of holes in the aeration tube.

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Figure 8. Design of solids drain.

Figure 9 shows the results obtained with rigid polyethylene aeration tubes containing various configurations of holes. (This tubing was actually microporous but previous testing had shown that the pores quickly plugged with coal fines and precipitates, so holes were drilled in it). In these experiments, a reactor section contained 3.5 lb of 20 x 40-mesh silica sand, 4.4 lb spent *T. ferrooxidans* medium to facilitate wetting of the coal, 12.2 lb coal, and sufficient water to fill the section. Sand was added before turning on the airflow. This slurry was aerated for 15 minutes at 2 SCFM after which one plug volume (the volume of the conical drain shown in Figure 8; 91 cm³) of solids was drained and returned to the section. After an additional 15 minutes aeration, another plug volume was drained, wet sieved to separate the sand, and the remaining solids were dried and analyzed for ash via ASTM Procedure D3174. This procedure was repeated at airflows down to 0.5 SCFM.

It was anticipated that more holes would generate a larger number of smaller bubbles and thus more efficient aeration, as well as a lower pressure drop through the aeration tube. The results (see Figure 9; the raw coal contained 9.55% ash) confirmed these expectations. The 12- and 20-hole tubes gave optimum separation at an airflow of 1.5 SCFM, while the 10-hole tube required a flow greater than 2 SCFM. The reason for the maximum separation is that at airflows less than 1.5 SCFM, there is insufficient turbulence to keep all the organic coal particles in suspension, so that they settle out along with the mineral particles. At higher flow rates there is so much turbulence that even the dense mineral particles stay in suspension, so separation is again reduced.

The data shown in Figures 10 and 11 for silicone rubber covered aeration tubes were obtained during the startup period when the aerated trough was run as a batch reactor and each section contained a 20% slurry of coal in a mixed culture of *Thiobacillus* and *Leptospirillum* bacteria, with no sand or glass beads. The pH had been equilibrated at 2.0 so that the basic minerals in the coal had dissolved, reducing its total ash content to approximately 6%, but leaving its insoluble, liberated ash (see Section 2.4) the same. Each sec-

tion had a slightly different aeration tube, although those in Sections 1, 2, and 4 were sufficiently similar (44, 40, and 38 holes respectively) to be considered identical. The airflows were set at the desired level and one plug volume of solids was drained and returned to the section. After 2 hours aeration, a further plug volume of settled solids was drained and analyzed for ash. For the 20-hole aeration tube in Section 3, the complete range of airflows was studied, with 15 minutes aeration being allowed at a given airflow rate before solids were drained and analyzed.

Figures 10 and 11, duplicate experiments done 12 days apart, show similar results. With 20 holes, gas dispersion was poor and there was no physical separation except at the highest airflow in Figure 10. Doubling the number of holes to the 38 to 44 range produced a sharp maximum in the physical separation at an airflow of 1.5 SCFM, exactly as in the previous experiment (see Figure 9). The drop between Aeration Tubes 4 and 5 was probably caused by a combination of fewer holes and lower airflow.

3.2.2 Operating Conditions

All the rigid aeration tubes were found to plug quickly. Typically one-half of the holes would be totally plugged after one week's operation, producing nonuniform aeration, higher pressure drops, and settling of coal over the plugged regions of the tube. Forty-hole silicone covered aeration tubes were therefore adopted. They resolved most of the difficulties of corrosion and settling experienced previously⁴ at the expense of a higher pressure drop, which translates into higher air compression costs. This pressure is needed to expand the tubing to open the holes to the point where air can flow out, and is not significantly reduced by increasing the number of holes (decreasing the tube wall thickness deserves investigation). At a constant airflow of 1.5 SCFM, the pressure would fall from 15 lb_f/in.² to around 8 lb_f/in.² over a period of weeks as the tubing fatigued and corrosion widened some of the holes. After approximately a month all the air would be flowing out of one or two large holes and the tube was replaced.

Figure 9. Results of physical separation study for porous polyethylene tubing.

1 = Polycarbonate 44 holes
 2 = Polycarbonate 38 holes
 3 = Acrylic 20 holes
 4 = Polycarbonate 40 holes
 5 = Acrylic 32 holes

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Figure 10. Results of physical separation study for silicone covered rigid tubing done on 11/15/91.

1 = Polycarbonate 44 holes
 2 = Polycarbonate 38 holes
 3 = Acrylic 20 holes
 4 = Polycarbonate 40 holes
 5 = Acrylic 32 holes

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Figure 11. Results of physical separation study for silicone covered rigid tubing done on 11/27/91.

Figures 9 through 11 show the importance of keeping the airflow at 1.5 SCFM in each section. This airflow is high enough to keep the organic particles in suspension, yet low enough to allow settling of the liberated minerals. This equals 1.4 on a gas volume per slurry volume per minute basis, and represents fairly intense aeration. It is three times that used previously⁴ and, based on that experience, oxygen transfer to the microorganisms should not be limiting even at the higher slurry concentrations. The corresponding air velocity (airflow/slurry surface area) is 1.4 ft/min. This velocity is proportional to the agitation power input per unit slurry volume and should be kept constant during scale-up in geometrically-similar reactors.

Values of the separation factor defined in Section 2.2 can be calculated from the data shown in this section at an airflow of 1.5 SCFM. It varies from 3.5 in Figure 10 to 2 in Figure 11; the difference arising because the bacteria were growing and generating biosurfactants between those two experiments, altering the gas dispersion and the hydrophobic interactions. The data in Figure 9, after correcting for the soluble basic minerals, suggest a similar value of $\alpha = 2.2$. This approximate value was used with Figures 2–6 from Section 2.2 to fix the initial flows of liquid and solids around the reactor.

3.3 Water Loss and Temperature Control

The combination of long liquid residence times and intense aeration in the reactor produces an obvious potential problem of evaporation. If the reactor is maintained at 24°C and the inlet air is totally dry, then the total airflow of 9 SCFM can evaporate up to 8.8 kg/d of water, which is on the order of one-third of the liquid feed rate. Such intense evaporation would alter the slurry concentration and its cooling effects need to be counteracted by considerable heating if the temperature is to be kept in the correct range for microbial metabolism.

In these experiments, the inlet air was humidified by a custom-built, steam-injection humidifier. It consisted of 11 ft of 1-1/8-in. diameter tubing giving an air residence time of 0.5 seconds. Steam was injected into the inflow through a metering needle valve, and excess condensate was removed through a trap at the outlet end. Tests showed that this consistently produced a humidity more than 80%. The addition of this humidifier facilitated temperature control by greatly reducing evaporative cooling. The temperature throughout the reactor could be maintained at $24.5 \pm 3.1^\circ\text{C}$ by three 100 W aquarium heaters located in Sections 1, 3, and 5.

It became apparent during the experiments that the majority of the water loss was due not to evaporation but to mist in the air leaving the reactor. This amounted to 6-7 kg/d (see Section 4.1). The loss caused by evaporation from the coal on the dewatering screen is unquantifiable and may contribute to this total. There was one demister/filter on the roof of each section except Section 2. Their design is shown in Figure 12. They were originally filled with glass wool, but this quickly became saturated and plugged, forcing the air to find alternative exits. This was replaced with a coarse plastic screen material, which drained well and would operate for several days between washings to remove accumulated coal fines (daily washings were needed with a 35% coal slurry in the reactor). Air demisting is another area requiring attention in future designs.

3.4 Startup, Operational, and Analytical Procedures

These experiments were done on the same coal as used previously.⁴ It was an Illinois #6 supplied by the Monterey Coal Company, with 9.55% ash, 4.04% total sulfur, 1.02% pyritic sulfur, a heating value of 12,700 Btu/lb, and a moisture content (after grinding) of 2.9%. The coal was ground to 80% -100 mesh at the Mountain States Energy (MSE) Inc. facility in Butte, Montana and shipped to the INEL under nitrogen in 55-gal drums.

Figure 12. Demisting device.

Details of the process startup procedure have been described previously.⁴ The reactor was filled with microbial medium containing 1.2 g/L Fe^{2+} as the energy source, and inoculated with a mixed culture of *Thiobacillus* and *Leptospirillum* from previous experiments. The medium was periodically recycled through an electrolytic cell to regenerate the Fe^{2+} and allow continued microbial growth. When the microbial cell concentration reached 1.4×10^9 cells per mL, coal was added. The resulting 20% slurry was aerated for 29 days in batch mode, and the coal and liquid feeds were turned on at the start of Day 30. An additional 30 days of continuous operation allowed the microbial culture to become properly adapted to the coal before any steady-state measurements were taken.

The daily procedures during the operating period were as follows:

3.4.1 Solids Drain

The solids were removed from the drains of Sections 1 through 6 on a daily basis. Since the conical drains (see Figure 8) could each hold a volume of approximately 100 mL and the drain strategy (see Figure 3) required removing a volume of 300-500 mL/section drain/day, the solids were removed repeatedly, allowing 20-30 minutes for the drains to refill with settled solids. After the initial solids were removed from each drain, we allowed a 20-30 minute waiting period for the drains to refill with settled solids. This procedure was repeated until 300-500 mL of solids was removed from each section. Note that the amount of solids that can be drained in this way is necessarily imprecise, but can be kept consistent when averaged over the reactor residence time (3-5 days). The solids drained from Sections 2-6 were wet sieved to remove the liquid, which was returned to the section from which it came. The sieved solids were weighed and added to Section 1.

The solids drained from Section 1 were also wet sieved, the liquid being distributed between Sections 2-6 to make up the volume lost with the drained solids. The sieved solids were weighed,

and samples taken for ash and (during certain steady-state periods) pyrite analysis.

3.4.2 Product Coal

The wet coal cake that had accumulated on the dewatering screen was weighed and spread on a drying tray. The wet coal was mixed and a representative sample was taken by filling a 500-mL beaker. The dry product coal was analyzed for total sulfur, heating value, pyritic sulfur, and ash. Before these measurements were taken, the coal was washed to remove the sulfate sulfur, which consisted of precipitate, sulfate sulfur in the coal, and that dissolved in the interstitial water. To measure sulfate sulfur and to prepare the coal for total sulfur and heating analysis, the following wash procedure was adopted. Approximately 30-40 g from the 500-mL beaker was added to 350 mL of 1.2 N HCl (10 mL 1.2 N HCl to 1.0 g of wet coal cake) and stirred for 30 minutes, then filtered. The solids were washed two more times following the same procedure. The filtrates were analyzed for iron by Atomic Absorption Spectroscopy and the sulfates by a barium chloride turbidimetric method (ASTM D-516-82). After the third wash, the extracted residue was air dried, then analyzed for ash, total sulfur, and heating value. MSE Inc., Butte, Montana performed the total sulfur and heating value analysis. To measure pyritic sulfur, 10 g of wet coal cake from the 500-mL beaker was dried, from which 5 g of dried coal was used to determine pyritic sulfur by ASTM D-2492-90.

The trays were allowed to dry in the laboratory for 10 days. They were then weighed and the coal on them mixed, and a representative sample was taken for analysis of the residual moisture. This allowed calculation of the masses of dry coal and moisture in the original cake.

3.4.3 Other Measurements

The wastewater reservoir was weighed and its contents mixed to suspend the coal fines. A 100-mL sample was taken with a graduated cylinder and allowed to settle in order to measure the fines concentration. A separate 4-mL sample was taken and acidified with 1 mL of 6 N HCl to

dissolve any precipitates. After at least 1 hour, the fines were filtered out and the filtrate analyzed for iron and sulfate as above. The wastewater reservoir was then emptied and replaced. During periods when the system was operated with no wastewater flow, samples were taken of the slurry from the reactor effluent, and analyzed in the same way. These measurements were used mainly to indicate when steady-state operation had been reached.

The weight of the feed reservoir was also noted. If necessary, it was refilled with tap water that had been passed through an activated carbon filter to remove any free chlorine. Ammonium

sulfate and 36 N sulfuric acid were also added at a rate of 0.49 g and 5.5 to 7.5 mL per kg of coal treated.

The pH, temperature, and oxidation/reduction potential (Eh) in each reactor section were also measured. Several checks were conducted, including the liquid flow rate into Section 1 and the reactor effluent slurry concentration (measured by collecting the effluent in a graduated cylinder and allowing it to settle overnight). Samples were also taken daily from the outlet reservoir and the reactor inlet liquid for direct microscope counts of total bacterial numbers.

4. RESULTS AND DISCUSSION

4.1 The Water Balance

The cumulative flows of water around the process during the 120 days of continuous operation are shown in Figure 13. The moisture in the feed coal is seen to be a small factor, increasing slowly during the experiment as the coal feed rate was increased. The water loss with the drained solids was always negligible. The dominant terms in the water balance are fresh liquid feed, the water in the wet product coal cake from the dewatering screen, the wastewater flow, and the loss by evaporation and misting [calculated from Equation (1)]. After Day 107, the wastewater flow was turned off to achieve the desired hydraulic and reactor residence times (τ_H and τ) and slurry concentration in Section 2 of the reactor (w).

Figure 13 shows that the flow rates (the slopes of the lines) were steady and consistent when averaged over the long time scales (the reactor residence times) involved in the experiment (the

discontinuity at Days 79 and 80 is caused by the flows not being measured on these days). However, the raw data (Table B-1, Appendix B) show that there was considerable variation between days. There are several reasons for this. The only flows that were actually fixed by pump controllers were the reactor inlet flow and the wastewater flow, both being calculated from the equations in Section 2 to give the desired values of τ_H , τ , and w . The occasional large variation in the other flows resulted from process upsets such as sticking of the reservoir float valve or plugging of the reactor outlet tube. Smaller fluctuations resulted from factors such as wear of the pump tubing and inevitable variations in the effectiveness of the dewatering screen from day to day. Another reason for the fluctuations was revealed by careful measurements of the slurry flow, made by collecting the reactor effluent for consecutive periods of 1 hour. These measurements showed flow surges as if the reactor was slowly overflowing over an 8–12 hour period and then returning to a lower level relatively quickly. These changes in level

Figure 13. The process water balance.

were not visible but, because of the large size of the reactor, they could involve a significant fraction of the daily flow (a 1 mm change in level in the reactor requires approximately 0.65 kg of slurry). These fluctuations will be eliminated in the future by changes in the design of the outlet tube and the holes in the baffles.

The most unexpected result from the water balance was the large loss of water from the process, which was an average of 7.8 kg/d over the 120 days of the experiment. It was originally thought that this loss was caused by evaporation. At the operating temperature of 24°C, the vapor pressure of water is 3 kPa, so at the airflow rate of 9 SCFM (254 L/min), the maximum possible evaporative loss (if inlet air is totally dry) is $3 \times 254 \times 60 \times 24 \times 0.018/101 \times 22.4 = 8.8$ kg/d. The letters A through F in Figure 13 indicate a progressive series of improvements to the inlet air humidifier, attempting to reduce this water loss. The humidifier reached the final form described in Section 3.3 at F (86 days) and measurements of inlet air humidity showed that it exceeded 80%, making the evaporation hypothesis untenable. In fact, the water loss was due mainly to mist droplets in the outlet air stream. The outlet air filters had reached the form described in Section 3.3 at E (72 days). They were originally filled with glass wool, which removed mist efficiently and, in conjunction with the air humidifier, reduced the average water loss to 6.1 kg/d between F and H. However, glass wool does not drain and quickly became saturated, leading to plugging and associated operating problems when the air sought other ways out of the reactor. At H, the glass wool was replaced with a larger plastic mesh (actually from plastic pan scrubbers). While this was slightly less efficient at mist removal (the slope of the Evaporation line in Figure 13 increases slightly) it drained more readily and led to fewer operational problems. The mesh was rinsed every few days to remove the accumulation of coal fines.

4.2 Solids and Heating Value Balances

The raw data on the flow rate of dry solids around the process is shown in Table B-2, and is

summarized on a cumulative basis in Figure 14. Once again, there is considerable daily variation, but the flows are steady over the time scale of interest. Nearly one ton of coal was treated over the 120 days of continuous reactor operation. The loss of coal fines with the wastewater was always negligible.

The process was operated under four different sets of operating conditions, the periods being marked by the vertical lines at 75, 107, and 132 days in Figures 13 and 14 (the line at 90 days marks a very slight increase in feed rate). At each condition, approximately four reactor residence times were allowed for the process to reach steady state. There followed a steady-state period of slightly over one residence time during which all readings were taken daily. These readings were averaged and summarized in Table 2. Note that the solids wasted from Section 1 were kept between 4% and 6% of the feed coal.

During the first two steady states, the mass of feed coal was closely matched by the sum of the product coal and the drained solids. This may seem unusual, since the dissolution of the basic ash and pyrite should reduce the total mass of solids. However, the product coal and solid waste were not washed before drying and weighing, so their measured weight included dissolved salts and precipitates from the interstitial water. This apparently canceled the loss of mineral matter. During the two final steady states, both at 35% slurry concentration, there was a definite loss of solids, amounting to approximately 1.6 kg/d. Some of this coal accumulated in the bioreactor; there was a noticeable increase in the deposition of solid clumps on the walls and lid of the reactor at the higher slurry concentration. Some coal also apparently left the reactor in the mist; the filters in the laboratory air conditioning system showed a surprisingly rapid accumulation of coal dust. It is also possible that some of the coal organic matter was being solubilized. After the experiments described here were completed and the system was returned to operating at $\tau = 7$ days and $w = 20\%$, a grab sample of slurry was taken from the reactor, filtered, and tested for dissolved carbon. It was found to contain 1,400 parts per million

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Figure 14. The process solids balance.

(ppm) of organic carbon suggesting that there was some solubilization of the coal, due either to chemical or microbial reactions. This merits further study, although it probably does not account for the observed loss of solids because there is no reason why it should be so much more intense in a 35% slurry than in a 20% slurry.

More important than the recovery of the mass of the feed coal is the recovery of its heating value. The heating value of the product coal, on a Btu/lb basis, shown in Table 2, was found to be consistently 5–7% less than that of the feed coal, despite its lower mineral content. This is consistent with previous results⁴ and may be caused by microbial oxidation of some part of the coal structure, which could also cause the solubilization discussed above. This loss, and the loss of heating value to the solid waste are combined in the heating value recovery defined by Equation (6). It is found to be in the range 80–90% for all the steady-state conditions (see Table 2).

4.3 Ash-Forming Minerals

The raw data on the ash content of the product coal and solid waste are given in Table B-3 and summarized as kilograms of ash per day in Figure 15. The ash flow with the product coal and the solid waste do not add up to that in the feed coal because some of the ash-forming minerals (including pyrite) are dissolved out of the coal during the process. Typically, 10% of the ash in the feed coal appeared in the solid waste, 60% appeared in the product coal, and 30% was solubilized. An ash removal of 40% in a process designed primarily to remove pyrite was an encouraging result, but the fact that most of it was dissolved rather than physically separated leaves considerable room for improvement. One way to assess the physical separation is the separation factor analysis given in Section 2. The data for the steady-state periods (see Table 2) show that the ash content of the solid waste was remarkably

Table 2. Steady-state process performance.

Days	Hydraulic residence time τ_H days	Reactor residence time τ days	Mass fraction coal in Section 2 (w)	Coal feed rate (dry) G_f (kg/d)	Product coal flow (dry) G_p (kg/d)	Drained solids D_1 (kg/d)	Ash in product coal A_6 (%)	Ash in solid waste (%)	Product heating value ΔH KJ/kg (Btu/lb)	Heating value recovery (%)
69-75	5.8	5	0.20	4.11	3.91	0.28	6.0	9.97	27,700 (11,900)	89
97-102	8.8	5	0.20	4.37	4.14	0.23	6.4	17.6	27,300 (11,800)	88
125-131	9.3	5	0.35	9.56	7.65	0.38	6.0	17.3	—	—
144-148	6.0	3	0.35	15.92	13.37	0.85	6.4	17.8	27,800 (12,000)	80

Feed coal values: Ash 9.55% (approximately 2.5% soluble basic minerals).
 ΔH 29,500 MJ/kg (12,700 Btu/lb).

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Figure 15. Ash removal.

constant at close to 17.6% (the lower value early in the experiment was probably caused by an inferior procedure for draining the solids). If it is considered that the 2% finely-dispersed, nonremovable ash assumed to be in the feed coal is still in this solid waste (see Section 2), this leaves 15.6% removable ash. Examination of Figure 4 shows that this corresponds to a separation factor $\alpha = 1.9$, close to the value of 2.2 determined in Section 3.3. The value $\alpha = 1.9$ was subsequently used, in conjunction with the Figures 2–6 in Section 2, to determine the feed rates and solids drain rates from the system.

The main reason for the low value of the separation factor is that Illinois #6 coals are difficult to clean physically. The float-sink curve for our particular coal, as ground, was not determined because of time and funding constraints, the diffi-

culties of the measurement for finely ground coal, and the fact that ash removal was not the primary objective of the process. Our estimate of 2% nonremovable ash may be conservative.

A second reason for the low separation factor concerns the mechanisms of physical separation. Mineral particles concentrate at the bottom of the trough because they are less hydrophobic and thus less likely to be carried upwards by the bubbles, and because of gravity settling. If gravitational effects dominate, then the critical quantity is the particle settling velocity. But settling velocity depends not only on the density, which is much higher for a mineral particle, but also on the particle size. A 150- μm organic coal particle has a similar settling velocity to a 50- μm liberated mineral particle, and thus a similar probability of appearing in the solid waste. To study this effect,

grab samples of feed coal, product, and solid waste were taken on Day 119 and subjected to particle size analysis on a Coulter Laser Particle Size Analyzer. The resulting particle volume distributions are shown in Figure 16. The feed coal had a mean size of 140 μm with a standard deviation of 105 μm . All the particles larger than 300 μm appeared in the waste stream, which had a mean of 214 μm and a standard deviation of 161 μm . The product was enriched in the fine particles with a mean of 109 μm and a standard deviation of 73 μm (although this may reflect reduction in particle size caused by mechanical abrasion and microbial activity, as well as the physical removal of the larger particles).

There appears to be considerable room for improvement in the physical separation performance of the process. The geometry of the reactor sections (particularly the depth) needs to be considered, and the arrangement of the aeration tube and solids drain optimized. Finally, a coal that contains more liberated ash and that has not been pre-cleaned (the fines stream from a coal preparation plant for example) would provide a better test of this aspect of the process.

4.4 pH, Eh, and Bacteria Counts

The oxidation/reduction potential (Eh) in microbial depyritization systems is a useful indicator of the ratio $\text{Fe}^{3+}/\text{Fe}^{2+}$ in solution. This ratio is decreased by the consumption of ferric ion for the chemical oxidation of pyrite:

It is increased by the subsequent oxidation of ferrous ion by the iron-oxidizing bacteria *Thiobacillus ferrooxidans* and *Leptospirillum ferrooxidans*.

A high Eh shows that there is sufficient microbial activity in the system to keep virtually all the dissolved iron in the Fe^{3+} state [i.e., to prevent the reaction presented in Equation (14) from becoming rate limiting].

The variation of Eh along the reactor during the four steady-state periods is shown in Figure 17. As expected, there is a sharp drop in Section 2 because the feed coal is added there, increasing the amount of fresh pyrite and accelerating the reaction presented in Equation (13). Thereafter, microbial iron oxidation dominates and the Eh increases along the reactor. The Eh also decreases with increasing slurry concentration and decreasing liquid recycle ($\tau_{\text{H}} \rightarrow \tau$). This is also as expected; less recycle means less Fe^{3+} in the reactor feed and more coal means that Fe^{3+} is consumed more rapidly by the reaction presented in Equation (13). Nevertheless, even under the most extreme conditions tested (35% slurry, $\tau = 3$ days, and $\tau_{\text{H}} = 6$ days), the Eh never fell below 500 mV, indicating that better than 90% of the dissolved iron was in the Fe^{3+} state.⁴

An unusual feature of the coal used in these experiments is that it contains a high ratio of basic mineral matter to pyrite (2.6% of the 9.55% total ash was found to be soluble in HCl). During microbial depyritization, the dissolution of these basic minerals produced a net increase in pH despite the acid formed by the oxidation (by catalyzed *Thiobacillus ferrooxidans* or *Thiobacillus thiooxidans*) of the sulfur generated by the reaction presented in Equation (13).

Note that much more acid could be generated by the formation of ferric hydroxysulfate precipitates, e.g., goethite,

However, such precipitates interfere with the process by coating the pyrite surfaces and complicating the washing of the product coal. A major process control objective is to keep the dissolved metal concentration and the pH within the tolerance of the microorganisms and low enough to prevent the formation of precipitates. The pH is kept within the desired range by adding sulfuric acid to the liquid feed reservoir. Note that there would be theoretical advantages to replacing this with phosphoric acid, which provides an essential

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Figure 16. Size distributions of feed, drain, and product coal.

Figure 17. Steady-state Eh values.

microbial nutrient. However, ferrous phosphate and other phosphates have very low solubilities, which would aggravate the formation of precipitates. The bacteria appear to obtain sufficient phosphate from the feedwater and coal.

The variation in pH along the reactor at the four steady-state conditions is shown in Figure 18. The sharp increase between Sections 1 and 2 was caused by the basic minerals in the feed coal. With a 35% slurry, the increase continued into Section 3, showing that the dissolution of these minerals was not instantaneous, or perhaps that there was an imbalance between the iron- and sulfur-oxidizing microbial activity in the system. Sulfur oxidation [reaction shown in Equation (15)] requires that the bacteria be attached to the pyrite surface, so many of the sulfur-oxidizing bacteria are lost from the system attached to the product coal. Iron oxidation [reaction shown in Equation (14)] can be catalyzed by bacteria in the liquid since Fe^{2+} , unlike S, is soluble. Thus, the recycled liquid probably contains a disproportionate number of iron-oxidizing bacteria and it takes time for the population of sulfur-oxidizing bacteria to grow to comparable levels. Since iron oxidation consumes acid [reaction shown in Equation (14)] while sulfur oxidation produces it [reaction shown in Equation (15)], this sequence of events may contribute to the rise in pH between Sections 2 and 3. In Sections 3 through 6, the pH drops steadily because of the microbial oxidation of pyritic sulfur.

In operating the process, the control objective was to add sufficient acid to the feed reservoir to keep the pH close to 2 throughout the reactor.

This pH is high enough to allow microbial metabolism and low enough to prevent the formation of precipitates. With a 35% slurry and $\tau_{11} = 9.3$ days, this strategy produced a sharp increase from pH = 1.84 to 2.14 between Sections 1 and 2. It is not known whether this sudden change affected microbial metabolism. However, this change is due solely to the high basic mineral content of the particular coal, and would not be a problem with most coals.

The Eh and pH provide excellent indirect measures of the health and activity of the microbial population. Obtaining more direct measures is difficult. Many of the bacteria attach to the coal making them almost impossible to enumerate. Also, it is difficult to distinguish between the various categories of bacteria; the autotrophic iron and sulfur oxidizers that oxidize the pyrite, the acidophilic heterotrophs that live symbiotically with them, and other heterotrophs that may be oxidizing the organic fraction of the coal. More research and testing are needed in this area. The only direct measure used in this experiment was direct microscope counts of the total bacterial concentration in the liquid in the reservoir under the product coal dewatering screen, and in the liquid fed to the reactor. The results are shown in Figure 19. The measurements are difficult and imprecise because of the presence of coal fines, and they show considerable daily variation around an average on the order of magnitude of 10^8 cells per mL. The liquid feed to the reactor consists of a mixture of feedwater and reservoir water, so the inlet data in Figure 19 is consistently lower than the reservoir data. The difference between them is consistent with the amount of liquid recycle, being high when the recycle ratio was low (Days 50–75) and low when recycle was high later in the experiment.

A disconcerting feature of the data in Figure 19 is that the cell counts appear to decline through the experiment as the coal feed rates to the process were increasing. This suggests that more cells attached to the product coal are lost from the process than can grow on the pyrite it contains, and demonstrates the importance of paying attention to the process cell balance as we seek to optimize the process by increasing the coal loading. Some important factors in this balance are as follows:

- Cell growth on the physically separated pyrite in Section 1 is essential
- Loss of pyrite in the solid waste stream means loss of cell production and may need to be reduced as the coal loading is increased

Figure 18. Steady-state pH values.

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Figure 19. Cell counts.

- Wastewater flow needs to be minimized to reduce cell loss.

Eliminating the wastewater flow from the process at Day 107 appeared to halt, temporarily, the decline in cell counts. There must also be a large loss of cells in the mist leaving the bioreactor with the outlet air, another reason for reducing this water loss. The overall process flow sheet⁴ includes a washing step for the product coal that was not studied in this experiment. The washing is designed to remove dissolved and precipitated sulfate from the wet product coal before it is burned, but it will also remove attached cells. Recycling this washwater into the process will make an important contribution to maintaining a high cell concentration.

Another important factor in the cell balance is maintaining the correct amounts of nutrients essential for cell growth. One nutrient of concern was the CO_2 needed for growth of the autotrophic bacteria.¹⁰ A grab sample of the slurry was therefore taken, filtered through a $0.2 \mu\text{m}$ filter and tested in a Total Organic Carbon Analyzer. It was found to contain 94 ppm of dissolved inorganic carbon, a surprisingly high number that is more than adequate to maintain microbial growth. The sample also contained 1,400 mg/L of organic carbon. The most likely explanation is that heterotrophic microorganisms are oxidizing and solubilizing part of the organic fraction of the coal, resulting in a slight loss of heating value (Section 4.2), a large increase in dissolved organic carbon, and high levels of dissolved carbon dioxide. The two important research questions are how to

minimize this activity, and why the high concentration of organics does not inhibit the autotrophic, pyrite-oxidizing bacteria that are normally extremely sensitive to dissolved organic matter.

One reason for doing daily microscope examinations of the microbial culture was to check that acidophilic protozoa did not appear in the system. These protozoa did appear in an adjacent experiment on the depyritization of coal in heaps. The protozoa devastated the performance of this process by grazing on the *Thiobacillus* and *Leptospirillum* responsible for pyrite oxidation. However, the protozoa never appeared in the aerated trough system, presumably because it could not survive the intense mechanical agitation and abrasion in the slurry. This is an important factor in the long-term stability of the proposed process.

4.5 The Sulfur Balance and Pyritic Sulfur Removal

The results of the daily sampling of the product coal during the four steady-state periods are shown in Table 3. Measurements of pyritic sulfur by the ASTM procedure, modified as described in Section 3.4, and of the total sulfur using a LECO analyzer were consistent during each period, showing that true steady states had been achieved. With a 20% slurry and 5 day reactor residence time, pyritic sulfur was reduced by 80% with a hydraulic residence time of $\tau_H = 5.8$ days (Days 69–75) and 87% with $\tau_H = 8.8$ days (Days 97–102). This confirms previous results⁴ that showed the benefits of liquid recycle and suggested that a recycle ratio around 1 (i.e., $\tau_H = 2\tau$) was best. This was adopted as the operating strategy for the remainder of the experiment.

For the $w = 20\%$ and $\tau = 5$ day conditions (the first two steady states in Table 3), total sulfur measurements confirmed the removal of pyritic sulfur. If the product coal contains the 3.02% organic sulfur in the feed coal and the 0.21% (at $\tau_H = 5.8$ days) or 0.13% (at $\tau_H = 8.8$ days) pyritic sulfur measured in the product, then the total sulfur

should be 3.23% and 3.15%, respectively. These are close to, although consistently higher than the measured values of 3.11% and 3.10% (note that the pyritic sulfur reduction rate calculated from the total sulfur measurements is consistently over 90%). However, at the most extreme conditions tested ($\tau = 3$ days and $w = 35\%$), the situation is very different. Although the pyritic and total sulfur measurements are both consistent through the steady-state period (Days 144–148), the pyrite measurement suggests a total sulfur of $3.02 + 0.38 = 3.40\%$, well below the measured value of 3.89%. The pyrite reduction would be 63% based on the pyritic sulfur measurements, but only 15% based on the total sulfur measurements. The most likely explanation for this involves the reaction mechanism discussed in the previous section. There is no reason to believe that the biodegradation of coal pyrite proceeds stoichiometrically; indeed there is experimental and theoretical evidence that it does not.¹¹ Iron oxidation [reaction shown in Equation (14)] and sulfur oxidation [reaction shown in Equation (15)] are catalyzed by two different sets of bacteria (although *T. ferrooxidans* can do both, it does not necessarily do both at the same time). The sulfur-oxidizing bacteria must be attached to the solid surface, which makes them more likely to be lost from the process with the product coal or the solid waste stream. Thus, even though the Eh data shows an active iron-oxidizing population, there may be a deficiency of sulfur-oxidizing bacteria. In conventional bioprocess engineering terminology, the mean cell residence time for the sulfur-oxidizing bacteria has fallen below the reciprocal of their specific growth rate and they have been washed out. The net result is that the elemental sulfur is produced by the reaction shown in Equation (13) faster than it can be oxidized by the reaction shown in Equation (15), and it accumulates in the system. It does not show up as pyritic sulfur because the ASTM procedure actually measures pyritic iron, the sulfur value being based on the stoichiometry of pyrite. However, elemental sulfur does appear in the total sulfur measurement, accounting for its high value. The solution is to maintain a higher population of sulfur-oxidizing bacteria by washing them off the product coal and returning them to the process.

Table 3. Measurements of product coal and solid waste.

Date	Day	Product coal		Btu/lb	Solid waste percent pyritic sulfur
		Percent total sulfur	Percent pyritic sulfur		
Raw coal	—	4.04	1.02	12,700	—
1/21/92	69	3.14	0.218	11,840	—
1/22/92	70	3.04	0.213	11,768	—
1/23/92	71				
1/24/92	72	3.08	0.205	11,481	—
1/25/92	73	3.19	0.210	12,039	—
1/26/92	74	3.07	0.212	12,183	—
1/27/92	75	3.15	0.203	12,162	—
	Average	3.11 ± .06	0.210 ± 0.005	11,912 ± 270	—
2/18/92	97	3.16	0.116	11,864	3.82
2/19/92	98	3.11	0.123	11,884	2.15
2/20/92	99	3.14	0.131	11,817	3.65
2/21/92	100	3.10	0.157	11,808	4.46
2/22/92	101	3.10	0.124	11,580	3.49
2/23/92	102	3.00	0.126	11,586	3.19
	Average	3.10 ± .06	0.130 ± 0.014	11,756 ± 137	3.46 ± .77
3/17/92	125	—	0.244	—	2.88
3/18/92	126	—	0.184	—	2.72
3/19/92	127	—	0.216	—	4.50
3/20/92	128	—	0.214	—	4.01
3/21/92	129	—	0.250	—	3.49
3/22/92	130	—	0.222	—	4.97
3/23/92	131	—	0.249	—	2.90
	Average		0.226 ± 0.024	—	3.64 ± .88
4/5/92	144	3.91	0.311	12,010	—
4/6/92	145	3.82	0.408	11,970	—
4/7/92	146	3.78	0.382	12,020	—
4/8/92	147	—	—	—	—
4/9/92	148	4.04	0.42	11,850	—
	Average	3.89 ± .12	0.380 ± 0.049	11,962 ± 78	—

An obvious question is the fraction of the pyrite reduction that is caused by microbial oxidation as opposed to physical separation. Since microbially-oxidized pyritic sulfur appears as sulfate in the liquid, this requires a complete sulfur balance for the process that evaluates all the terms in Equation (3). This is shown in Table 4, all terms being in grams of sulfur per day, averaged over the steady-state periods. The sulfate sulfur in the feed coal and the solid waste were found to be negligible and are not included. Summing the sulfur outflow in the wastewater, the product coal (pyritic and sulfate), and the drained solids (pyritic) and comparing this result with the total sulfur inflow always showed a large discrepancy. One reason for this discrepancy must be the loss of sulfate with the mist in the air leaving the reactor, a phenomenon whose significance had not been anticipated during planning of the experiment. Its magnitude was estimated from the known water loss rate and average sulfate concentration in the reactor and included in Table 4. The addition of this term closed the process sulfur balance within the expected limits of error ($\pm 5\%$) for the first two steady states. However, in the last two steady states, where the slurry concentration was 35% rather than 20%, a significant fraction ($\sim 20\%$) of the sulfur fed into the process remains unaccounted for. Some of this apparent sulfur loss may be caused by the accumulation of solids on the walls and roof of the reactor, which was unavoidable with these concentrated slurries (see Section 4.2). However, most of the sulfur loss must be caused by the partial oxidation of pyrite discussed above. Some of the pyritic sulfur in the feed is converted to elemental sulfur rather than sulfate and does not appear in any of the outflow measurements. This hypothesis is supported by the pyritic and total sulfur measurements from the fourth steady state (see Table 3), which suggests that the product coal contains $0.49 \pm 0.17\%$ elemental sulfur, corresponding to a sulfur flow of between 42.8 and 88.2 g/d. The low end of this range is the amount needed to close the process sulfur balance. During the second and third steady-state periods, the pyrite in the solid waste was measured directly. It was found to be approximately 3-1/2 times that in the feed coal, corresponding to physical removals of 14.3% and 17.7% (note that the drained pyrite numbers in

Table 4 for the first and last steady states, when these measurements were not made, come from assuming an average value of 16% removal). This is a similar separation performance as was found from ash measurements (Section 4.2) and is not a good result from a two-stage process. This is partly caused by the nature of the Illinois #6 coal, which is known to be difficult to clean physically. However, it must not be forgotten that the aerated trough was originally designed purely as a bio-reactor, and has not been optimized for physical separation. Further development, particularly changes in the solids drain and increases in the slurry depth, should give much improved physical removal of both pyrite and ash.

The percent removal of pyritic sulfur caused by microbial oxidation is calculated from the data in Table 4 for the rate of appearance of sulfate (i.e., the first form of Equation (5); the second form is not valid when elemental sulfur is being produced). As expected, the percent oxidation decreases with decreased reactor residence time and increased coal slurry concentration, from 65–70% with $\tau = 5$ days and $w = 20\%$ to 26% with $\tau = 3$ days and $w = 35\%$. This raises the question of the combination of physical separation and microbial oxidation that would work best in practice. Higher levels of microbial oxidation require larger reactors. On the other hand, there is a need to biodegrade significant amounts of pyrite to grow enough sulfur oxidizing bacteria to keep the process going. A process that approaches complete pyrite oxidation also has an environmental advantage: the solid waste it produces can no longer produce any acid drainage. These competing requirements may best be met by combinations of physical separation and biodepyritization other than that shown in Figure 1. One can, for example, envision a 7-stage reactor, with the coal fed into Section 4, the solids drained from Sections 4–7 going to Section 2, and those drained from Sections 2 and 3 going to 1. The focus should not be on the overall reactor residence time, but on the time different particles spend in the reactor. A pure, organic coal particle will pass through quickly, while a liberated pyritic inclusion is trapped in the system for several days by the mechanisms of physical separation.

Table 4. The process sulfur balance.

Steady-state conditions			Sulfur inflow (g/day)			Sulfur outflow (g/day)				Percent removal		
τ days	τ_H days	w percent	Coal pyrite	Feed reservoir	Waste- water	Product pyrite	Product sulfate	Drained pyrite	Sulfate in mist	Total	Physical	Microbial oxidation
			$G_f X_f$	$F_f S_f/3$	$F_w S_w/3$	$G_p X_p$	$G_p X_p$	$D_1 G_1 X_1$	$ES/3^a$		$D_1 G_1 X_1$	
5	5.8	20	1.9	15.2	21.6	8.6	13.2	6.7 ^b	9.9	60.0	16 ^b	70.4
5	7.3	20	44.6	13.5	13.5	5.5	19.1	7.9	10.1	56.1	17.7	65.5
5	8.8	35	97.5	30.7	0	17.2	44.1	13.9	25.0	100.2	14.3	39.4
3	6.0	35	162.4	61.5	0	50.7	76.0	26.0 ^b	28.4	181.1	16 ^b	26.4

a. Estimated based on known water loss rate and average sulfate concentration in reactor.

b. Not measured; estimated based on 16% physical pyrite removal.

This work produced several important conclusions about the need for caution in interpreting the results of standard analytical procedures for microbial coal depyritization. The literature contains claims for very rapid apparent microbial depyritization, based on the disappearance of pyritic sulfur. Some of these are from slurry bioreactor systems and are caused by physical settling of the large pyrite inclusions. Other claims overlook the possibility of partial oxidation of pyrite to elemental sulfur and the fact that the ASTM pyrite procedure actually measures pyritic iron. Separate measurements of total or elemental sulfur in the coal or sulfate sulfur generated are essential to confirm the removal of pyritic sulfur. It was also found that the ASTM procedure for removing non-pyritic iron by boiling in HCl before a pyrite measurement is inadequate for samples containing ferrie hydroxysulfate precipitates generated by microbial activity. Repeated washings are neces-

sary to make the pyrite measurement accurate. An example of the errors that can arise is given in Figure 20. It seemed reasonable to suppose that the amount of sulfate in the product coal removed daily from the dewatering screen could be found by multiplying the amount of water it contained by the concentration of sulfate in the liquid in the slurry leaving the reactor ($\square$ in Figure 20). This effluent slurry appeared to contain no precipitates because a standard procedure adding 1 mL of 6 N HCl to 4 mL of slurry, mixing for 1 hour, filtering and measuring sulfate in the filtrate produced no increase in the measurements ($*$ in Figure 20). However, a direct measure of the sulfate in the wet coal cake using repeated washings in 1.2 N HCl, consistently produced values that were 2 or 3 times higher ($\diamond$ in Figure 20). Some of the difference can be explained by evaporation of water from the coal cake on the dewatering screen. Some of the difference, however, seems to be because the

Figure 20. Sulfate in product coal.

slurry in the reactor effluent does contain some precipitates that were not dissolved and detected by the standard procedure described above. Some of the difference remains unexplained; the values in Table 4 are the higher values determined by direct measurement.

4.6 The Process Iron Balance

The interpretation of the data in the previous sections is confirmed by measurements of the forms of iron entering and leaving the process. The data are given in Table B-4 and the averages over the steady-state periods are summarized in Table 5. Iron enters the process as pyrite in the feed coal and leaves as (a) pyrite in the product coal and solid waste, (b) dissolved and precipitated iron in the water associated with the product coal, (c) iron dissolved in the wastewater, and (d) iron in the mist leaving the reactor with the air. The solid waste flow for the first and last steady states, and the mist loss are estimated as for the sulfur balance. The results show a slight surplus of outflow over inflow for the first three steady states and a deficit for the last steady state that is less than expected from the sulfur and total solids balances (because of accumulation of solids on the walls and roof of the reactor). The most probable explanation for the imbalance is the presence of some nonpyritic iron (e.g., FeCO_3) in the soluble basic mineral fraction of the feed coal.

The percent removal of iron by physical separation is not shown in Table 5, but must be identical to the physical removal of pyritic sulfur in Table 4. Both are calculated from the pyrite measurements in the feed coal and solid waste where pyrite can safely be assumed to be FeS_2 . More significant is the comparison between the percentage of the pyritic sulfur oxidized to sulfate (Table 4) and the percentage of the pyritic iron that is solubilized and appears in one of the liquid effluents (Table 5). At a 20% slurry concentration, the iron solubilized is very similar to the sulfur oxidized: the slightly higher values for iron (74 and 81% vs. 70 and 66%) probably being caused by the presence of nonpyritic iron in the feed coal. Furthermore, adding the percent sulfur removals by physical and microbial means gives values (86 and 82%) con-

sistent with the percent reductions calculated from the pyritic sulfur measurements $[(1.02-.21)/1.02 = 79\% \text{ and } (1.02-.13)/1.02 = 87\%]$ and independently from the total sulfur measurements in Table 3 $[(4.04-3.11)/(4.04-3.02) = 91\% \text{ and } (4.04-3.10)/(4.04-3.02) = 92\%]$. It appears that the process is working well; there are plenty of both iron- and sulfur-oxidizing bacteria in the system, and the pyrite is being solubilized stoichiometrically. The rate limiting step is probably the purely chemical reaction shown in Equation (13).

Contrast this with what happens when the coal loading is increased ($w = 35\%$, $\tau = 5$ days, and 3 days). Both the percent sulfur oxidized and the percent iron oxidized are reduced, as expected, but the iron removal is now much larger (70 and 51% for iron vs. 39 and 26% for sulfur). This is again consistent with the numbers for percent reductions. The pyrite measurements for the feed and product, which are actually measurements of pyritic iron, give solutions of $(1.02-.226)/1.02 = 78\%$ and $(1.02-.38)/1.02 = 63\%$, which agree reasonably well with the total removals obtained by summing the iron solubilization and physical removal effects (84 and 69%). However, the total sulfur measurements for $\tau = 3$ days (not available for $\tau = 5$ days) give a reduction of only $(4.04-3.89)/(4.04-3.02) = 15\%$. What these numbers suggest is that at the higher coal flows, sulfur-oxidizing bacteria attached to the product coal are being removed from the system faster than they can grow. The reaction shown in Equation (15) becomes rate limiting and a passivating layer of elemental sulfur forms on the pyrite surface, slowing both iron and sulfur solubilization as shown by the lower removals in Tables 4 and 5. However, sulfur oxidation is more seriously affected because more iron than sulfur has been solubilized, which accounts for the values in Table 5 being higher than those in Table 4. The accumulated elemental sulfur does not show up in the ASTM pyrite measurements, which actually measure pyritic iron. However, it does show up in the total sulfur measurements, which accounts for the much lower reductions calculated from these measurements. The consistency of values calculated (such as pyrite reduction) from independent measurements of dissolved iron, dissolved sulfate, pyrite, and total sulfur is a useful test of the data.

Table 5. The process iron balance.

Table 3. The process iron balance.										
τ (d)	w percent	τ_H (d)	Feed coal g Fe/d	Out: gram Fe/d						
				Product coal liquid	Product coal pyrite	Waste water	Drained solids	Mist ^b loss	Total	Percent Fe solubilized ^c
5	20	5.8	36.5	9.4	7.5	13.0	5.8 ^a	4.6	40.3	74
5	20	7.3	38.8	15.7	4.7	8.5	6.9	7.2	43.0	81
5	35	8.8	84.8	46.5	15.0	0	12.1	12.7	86.3	70
3	35	6.0	141.4	55.1	44.2	0	22.6 ^a	16.3	138.2	51

a. Not measured: based on 16% removal.

b. Estimated based on mist loss rate and average dissolved iron concentration in reactor.

c. (Product coal liquid + wastewater + mist loss)/feed coal.

a. Not measured: based on 16% removal.

b. Estimated based on mist loss rate and average dissolved iron concentration in reactor.

c. (Product coal liquid + wastewater + mist loss)/feed coal.

In summary, it is difficult to give a single figure for pyrite removal because pyrite does not necessarily remain pyrite during the process. The best numbers are the reductions based on total sulfur measurements calculated above, and the physical and microbial removals shown in Table 4. The latter removals do not add up to the former because reduction only compares the quality of the product and the feed coals, while a removal is calculated from the flows of sulfur in feed and

product. These measures show that the goal of 90% removal of pyritic sulfur was reliably achieved with a slurry concentration of $w = 20\%$ and a reactor residence time of $\tau = 5$ days. Increasing the coal loading by increasing w or reducing τ reduced the removal primarily caused by the loss of sulfur-oxidizing bacteria. Changes in process configuration are expected to resolve this problem and achieve 90% reduction at higher loadings.

5. CONCLUSIONS AND RECOMMENDATIONS

5.1 Conclusions

This work has given a first demonstration of the possibilities of combining physical and microbial processes for removing pyrite from coal. The process is based on two stages of physical separation, with the rejected solids from the first stage being transferred to the first reactor section, which acts as a rougher. Bacterial growth in this section, initiated by bacteria in the liquid recycle stream, serves to inoculate the feed coal (added to the second section) and maintain a large, active cell population. The objective is to physically remove the large pyritic inclusions, which would take too long to biodegrade, while biodegrading the many small inclusions, which are difficult to separate physically without very fine grinding. Theoretical considerations, based on typical size distributions of pyritic inclusions, suggest that 90% of the pyrite could be removed from a coal ground to -100 mesh with a residence time of 1 day.

Considerable progress was made towards demonstrating this performance in a large-laboratory scale (150-L slurry volume) process. The reactor was operated continuously for 120 days, treating approximately 1 ton of an Illinois #6 coal. Ninety percent pyrite removal was achieved at a 20% slurry concentration and a reactor residence time of 5 days; 16% attributed to physical pyrite separation and the remainder to microbial oxidation. This was accompanied by 40% ash removal; approximately 30% being caused by dissolution of basic minerals and 10% to physical separation. Ninety percent of the heating value of the feed coal was recovered; the loss being divided about equally between slight oxidation of the coal organic matter and loss of heating value with the solid waste from the physical separation.

At a three day residence time and 35% slurry, heating value recovery dropped to 80% (although the heating value per pound of product had increased) and pyrite measurements using the ASTM Procedure D-2492-90 suggested a pyrite

reduction of over 60%. However, this is actually a measurement of pyritic iron, and some of the sulfur was actually converted to elemental form rather than being oxidized to sulfate. This problem can be overcome by taking measures to retain sulfur-oxidizing bacteria in the process. These measures include incorporating washing of the product coal into the process and reducing mist losses from the reactor. By combining these measures with improvements in the physical separation process, further development should allow the process to meet the theoretical expectations with many coals.

The experiments reported here demonstrated solutions to many of the operational problems identified in previous work, and extended the results to higher coal loadings. The silicone rubber aeration tubes gave uniform aeration and, with the correct airflow rate, kept the organic fraction of the coal in suspension while allowing the denser mineral particles to settle out. They also corroded far more slowly than previous designs. The importance of liquid recycle was confirmed, and a recycle ratio close to 1 was again found to give the best compromise between the need to inoculate the feed coal with bacteria and ferric iron, and the effect of high concentrations of inhibitory products that can accumulate around the recycle loop. When operating at this recycle ratio and at high slurry concentrations, a commercial process would generate relatively little, concentrated wastewater that would be treated and either recycled to the process or discarded and made up by clean water. The clean or recycled water would be used first for washing the product coal and the wash effluent, with all of its valuable bacteria, fed into the process. The wastewater treatment (probably by lime precipitation) and coal washing steps are essential parts of the process that must be studied in future work. The present laboratory-scale process actually generated no wastewater, but this is partially due to the high "mist" losses which must be eliminated in the future.

5.2 Recommendations

Research should continue to find optimum values of the main process variables of reactor residence time, slurry concentration, and process hydraulic residence time (or recycle ratio). The objective must be to maximize the amount of coal that can be processed per unit reactor volume per unit time [proportional to (slurry concentration)/(reactor residence time)] with the desired level of pyritic sulfur removal. The main difficulty is simply that of time. In a process with hydraulic residence times of a few days, stable operation for a few weeks is required to achieve a single steady state.

The geometry of the individual reactor sections needs to be modified to optimize the physical separation of liberated pyrite and other insoluble minerals. This separation is caused by a combination of gravity settling and hydrophobic interactions with air bubbles, and would presumably be enhanced in a deeper, narrower vessel. Column flotation of coal may provide some guidelines, although there are significant differences because of the absence of froth. Considerable data exists in the literature both on the effect of *Thiobacillus* bacteria and the biosurfactants they produce on the flotation process, and on the possibly inhibitory effects of conventional frothers on the bacterial metabolism. However, much of this information is contradictory and no firm conclusions can be drawn at present.

Demisting of the air leaving the process must be improved. The appearance of an acidic mist containing bacteria and coal fines is an environmental problem that could inhibit process development. Also, the ability to maintain a high concentration of viable bacteria at high coal loadings is adversely affected by loss of cells in the mist. In thinking about changes to the shape or size of a slurry reactor, the quantity that must be kept constant (as a first approximation) is the agitation power input per unit slurry volume. This is proportional to the product of the slurry depth (i.e., the liquid head) and the airflow rate per unit volume, a product that equals the superficial air velocity defined in Section 3.2.2. It follows that scaling up the

existing process simply by increasing the depth has several advantages. The physical separation of coal from mineral matter can reasonably be expected to improve. Also, the airflow rate, and thus, the mist losses and the air compressor power needed to overcome the pressure drop in the aeration tubes, remain constant while the amount of coal being processed increases. When changing the shape or size of a slurry reactor, the quantity that must be kept constant is the agitation power input per unit volume. This is proportioned to the product of the reactor depth (i.e., the liquid head) and the airflow rate per unit slurry volume, a product that equals the superficial air velocity defined in Section 3.2.2. It follows that scaling up by increasing reactor depth has several advantages. Not only can physical separation be expected to improve, but the airflow rate, the mist losses, and the air compressor energy needed to overcome the pressure drop in the aeration tubes all remain the same while the amount of coal being processed increases.

Coal contains relatively little pyrite, and the growth yield of bacteria on pyrite is small. It follows that maintaining a high cell concentration in the system requires active measures to conserve cells. Besides controlling the liquid recycle and solids wastage, and reducing the loss caused by misting requires that the product coal be washed, with the washwater recycled to the process. Future testing would best be done on a coal that has not been cleaned and contains higher levels of pyrite and ash. The fines discarded by many coal preparation plants would be ideal for testing.

Methods are needed to separate and quantify the different classes of microbial activity: iron oxidation, sulfur oxidation, heterotrophic activity that degrades inhibitors of iron and sulfur oxidation (beneficial), and heterotrophic activity that solubilizes and oxidizes coal (harmful). The data show that at the highest coal loading studied ($\tau = 3$ days and $w = 35\%$), the loss of heating value was reduced but iron oxidation exceeded sulfur oxidation activity. Controlling and balancing these categories of microbial activity requires that they first be quantifiable in a coal slurry.

Considerable care is needed when applying ASTM procedures to microbial depyritization processes. The preliminary boiling in HCl will not remove all the nonpyritic iron from a coal that has been in a slurry containing high levels of ferric ion and ferric hydroxysulfate precipitates. It should be preceded by soaking the coal in a large

volume of dilute HCl. It must also be noted that the procedure actually measures pyritic iron, and that microbial degradation of coal pyrite does not necessarily proceed stoichiometrically. Pyritic sulfur may be changed to elemental sulfur, which does not show up in the ASTM procedure, but will appear in a total sulfur analysis.

6. REFERENCES

1. L. L. Josephs and D. D. Ferris, "Engineering Development of Advanced Froth Flotation—Pyrite Liberation Study and Round–Robin Flotation Report," *Proceedings 6th Annual Coal Preparation Utilization and Environmental Control Contractors Conference*, Pittsburgh, Pennsylvania, August 1990, p. 298–304.
2. G. F. Andrews, C. J. Stevens, and J. E. Wey, "The Log–Normal Distribution of Pyritic Inclusions in Coal, Consequences for Depyritization," *3rd International Symposium on the Bioprocessing of Coal*, DOE/EPRI, Clearwater, FL, May 1992.
3. S. McCaughey and M. McIlwain, *Interim Conceptual Design for a Commercial–Scale Coal Biodesulfurization Plant*, MSE, Inc., Butte, MT, August 1992.
4. G. F. Andrews, P. R. Dugan, M. E. McIlwain, and C. J. Stevens, *Microbial Desulfurization of Coal*, EG&G–2669, March 1992.
5. G. F. Andrews, P. R. Dugan, and C. J. Stevens, "Combining Physical and Bacterial Treatment for Removing Pyritic Sulfur from Coal," *In Processing and Utilization of High–Sulfur Coals*, Vol. IV, P. R. Dugan, D. Quigley and Y. Attia (eds), New York: Elsevier Publishers, 1991, pp. 515–531.
6. G. F. Andrews, "Mixing Effects in Coal Processing Bioreactors," *Resources Conservation and Recycling* 3, 1990, pp. 137–148.
7. Y. A. Attia, "Feasibility of Selective Biomodification of Pyrite Floatability in Coal Desulfurization by Froth Flotation: Effect of Bacterial Adaption," *Proceedings 3rd DOE Bioprocessing of Coals Workshop*, Tyson's Corner, VA, August 1988, pp. 199–209.
8. G. Rossi, "Microbial Depyritization of Coal," *3rd International DOE/EPRI Symposium on the Bioprocessing of Coal*, Clearwater, FL, May 1992.
9. G. F. Andrews and J. Quintana, "Mixing and Mass Transfer in the Aerated Trough Bioreactor," *Proceedings 1st International Symposium on the Biological Processing of Coal*, Orlando, Florida, EPRI GS6970, 1990, pp. 5–69–5–85.
10. G. F. Andrews, M. Darroch, and T. Hanson, "Bacterial Removal of Pyrite from Concentrated Coal Slurries," *Biotechnology & Bioengineering*, Vol 32, 1988, pp. 813–820.
11. G. F. Andrews, "An examination of the Kinetics of Coal Pyrite Decomposition," *In: Biotechnology in Materials and Metal Processing*, B. Scheiner, F. Doyle and S. Kawatra (eds), Society of Mining Engineers, 1989, pp. 87–93.

Appendix A

Solution To The Ash Removal Equations

Appendix A

Solution To The Ash Removal Equations

Equation (10) can be thought of as the drain strategy equation because it defines the process parameter β . As written it applies to any section and it can be solved for D_i :

$$D_i = \frac{A_i G_i \beta}{1 - A_i \alpha \beta} = B_i G_i \beta \quad (\text{A-1})$$

where B is defined as $A/(1-\beta\alpha A)$. The first step in the solution is to use this equation to eliminate the six unknown D values from the mass balance Equations (7), (8), and (9). For $3 \leq i \leq 6$ eliminating D_i from the first Equation (7):

$$g_{i-1} = g_i(1 + \beta B_i) \quad (\text{A-2})$$

where

$$g_i = G_i/G_f$$

Eliminating D_i and G_i from the second Equation (7) using (A-1) and (A-2) gives

$$A_{i-1} = \frac{A_i(1 + \alpha\beta B_i)}{1 + B_i\beta} = \frac{B_i}{1 + B_i\beta} \quad (\text{A-3})$$

where the second equality follows from the definition of B .

For Section 2 the same procedure involving Equations (A-1) and (8) gives:

$$g_1 = g_2(1 + \beta B_2) \quad (\text{A-4})$$

$$g_1 A_1 = g_2 B_2 - A_f \quad (\text{A-5})$$

Now take Equation (A-1) applied to Section 1 and substitute in the first Equation (9):

$$g_6 = 1 - \beta g_1 B_1 \quad (\text{A-6})$$

$$g_6 = \frac{A_f - \alpha A_1}{A_6 - \alpha A_1} \quad (\text{A-7})$$

Equations (A-2) to (A-7) are now twelve equations in the twelve unknowns A_i, g_i for $i=1$ through 6. It is actually simpler to specify a desired product

quality, A_6 (along with values of α and β), and to treat the feed composition A_f as an unknown. Equations (A-2) and (A-3) can then be solved for A_2, A_3, A_4, A_5 and the ratios $g_5/g_6, g_4/g_5, g_3/g_4$, and g_2/g_3 , leaving (A-4) to (A-7) as four equations in four unknowns A_1, A_f, g_6 , and g_1 . Equations (A-4) to (A-7) actually reduce to a quadratic equation for A_1 .

Converting g_2 in Equation (A-5) with the ratio $b=g_2/g_6$, and substituting Equation (A-7) for g_6 yields

$$g_1 A_1 = b B_2 \frac{(A_f - \alpha A_1)}{(A_6 - \alpha A_1)} - A_f \quad (\text{A-8})$$

Equating Equation (A-6) to (A-7) yields

$$g_1 A_1 = \frac{(A_6 - A_f)}{(A_6 - \alpha A_1)} \left(\frac{1}{\beta} - \alpha A_1 \right) \quad (\text{A-9})$$

Setting Equation (A-8) equal to Equation (A-9) and solving for A_f yields

$$A_f = \frac{A_6 \frac{1}{\beta} - \alpha A_1 + \alpha A_1 b B_2}{b B_2 - A_6 + \frac{1}{\beta}} \quad (\text{A-10})$$

Dividing Equation (A-5) by Equation (A-4), substituting Equation (A-7) for g_6 and solving for A_f yields

$$A_f = \frac{\alpha A_1^2 [b(1 + \beta B_2) - 1] + A_1(A_6 - b B_2)}{A_1[b(1 + \beta B_2) - \alpha] + A_6 - b B_2} \quad (\text{A-11})$$

Equating Equation (A-10) with (A-11) and cross multiplying yields

$$\alpha A_1^2 \left[\frac{b-1}{\beta} + A_6(1 - \alpha) + \alpha b B_2 \right] + \frac{A_1}{\beta} [A_6(A_1\beta(\alpha - 1) + 1 + \alpha - b) - \alpha b B_2(1 + A_6\beta\alpha)] - \frac{A_6}{\beta} (A_6 - b B_2) = 0 \quad (\text{A-12})$$

Once A_1 is solved from Equation (A-12), the feed composition is then found from Equation (A-10). Values of g_6 and g_1 can then be found from Equations (A-7) and (A-4) respectively.

Appendix B

Raw Data

Table B-1. The water balance.

Table B-1. The water balance.													
Day	Fresh feed (kg/d)	Feed coal			Drain		Product coal			Wastewater			
		Wet (kg/d)	H ₂ O (kg/d)	H ₂ O (kg/d)	Wet (kg/d)	H ₂ O (kg/d)	Wet (kg/d)	Moisture (%)	H ₂ O (kg/d)	Total (kg/d)	Volume settled solids/total volume	Mass fraction	H ₂ O (kg/d)
30	23.38	4.23	0.12	0.68	0.24	3.925	44	1.73	8.278	0.010	0.005	8.24	
31	18.97	4.23	0.12	0.09	0.03	3.287	44	1.45	6.603	—	0.005	6.57	
32	20.53	4.23	0.12	0.09	0.03	4.18	44	1.84	8.632	0.012	0.006	8.58	
33	21.53	4.23	0.12	0.09	0.03	4.154	44	1.83	8.07	0.008	0.004	8.04	
34	21.17	4.23	0.12	0.23	0.08	5.208	44	2.29	8.865	0.011	0.005	8.82	
35	20.46	4.23	0.12	0.314	0.11	5.897	44	2.59	8.707	0.032	0.015	8.57	
36	22.05	4.23	0.12	0.234	0.08	5.865	44	2.58	9.421	0.016	0.008	9.35	
37	23.41	4.23	0.12	0.18	0.06	5.803	44	2.55	10.329	0.010	0.005	10.28	
38	23.29	4.23	0.12	0.216	0.08	6.495	24.3	1.58	10.01	0.016	0.007	9.94	
39	23.91	4.23	0.12	0.3	0.11	6.9	26.6	1.84	9.627	0.011	0.005	9.58	
40	22.55	4.23	0.12	0.318	0.11	5.47	29.3	1.60	9.154	—	0.005	9.11	
41	19.28	4.23	0.12	0.326	0.11	7.923	24.9	1.97	9.674	0.012	0.006	9.62	
42	22.57	4.23	0.12	0.328	0.11	6.463	34.2	2.21	9.852	0.013	0.006	9.79	
43	21.72	4.23	0.12	0.534	0.19	6.444	36.6	2.36	12.968	0.014	0.006	12.89	
44	21.95	4.23	0.12	0.492	0.17	6.292	32.9	2.07	9.407	0.013	0.006	9.35	
45	22.14	4.23	0.12	0.472	0.17	5.758	36.4	2.10	9.603	0.012	0.005	9.55	
46	23.24	4.23	0.12	0.542	0.19	4.423	45.6	2.02	9.933	0.005	0.002	9.91	
47	22.52	4.23	0.12	0.442	0.15	5.388	41.1	2.21	9.42	0.021	0.010	9.33	

Table B-1. (continued).

Table B-1. (continued).

Day	Fresh feed (kg/d)	Feed coal			Drain			Product coal			Wastewater			
		Wet (kg/d)	H ₂ O (kg/d)	Volume settled solids/ total volume	Wet (kg/d)	H ₂ O (kg/d)	Moisture (%)	Wet (kg/d)	H ₂ O (kg/d)	Total (kg/d)	Mass fraction	H ₂ O (kg/d)		
47	22.52	4.23	0.12	0.442	0.15	5.388	41.1	2.21	9.42	0.021	0.010	9.33		
48	19.78	4.23	0.12	0.344	0.12	6.382	40	2.55	8.413	0.010	0.005	8.37		
49	21.56	4.23	0.12	0.52	0.18	6.694	31.2	2.09	8.697	0.011	0.005	8.65		
50	21.95	4.23	0.12	0.568	0.20	7.408	27.9	2.07	8.787	0.011	0.005	8.74		
51	20.56	4.23	0.12	0.546	0.19	7.939	31.87	2.53	9.47	0.011	0.005	9.42		
52	20.16	4.23	0.12	0.34	0.12	8.018	44	3.53	9.989	0.011	0.005	9.94		
53	10.37	4.23	0.12	0.334	0.12	7.909	44	3.48	0	—	0.005	0.00		
54	23.08	4.23	0.12	0.468	0.16	6.846	43.07	2.95	9.658	0.020	0.010	9.57		
55	19.60	4.23	0.12	0.534	0.19	7.934	30	2.38	9.84	0.020	0.010	9.75		
56	17.85	4.23	0.12	0.384	0.13	8.288	25.7	2.13	9.602	0.010	0.005	9.56		
57	20.39	4.23	0.12	0.41	0.14	5.609	27.9	1.56	7.636	0.082	0.039	7.34		
58	20.10	4.23	0.12	0.542	0.19	6.932	44	3.05	9.738	0.026	0.012	9.62		
59	21.11	4.23	0.12	0.31	0.11	6.232	44	2.74	9.252	0.011	0.005	9.20		
60	18.35	4.23	0.12	0.36	0.13	7.762	44	3.42	9.292	0.012	0.006	9.24		
61	19.58	4.23	0.12	0.402	0.14	7.111	29.45	2.09	9.395	0.011	0.005	9.35		
62	19.68	4.23	0.12	0.6	0.21	6.883	34	2.34	9.379	0.073	0.034	9.06		
63	20.60	4.23	0.12	0.252	0.09	7.123	36.7	2.61	9.545	0.005	0.002	9.52		
64	17.71	4.23	0.12	0.35	0.12	6.165	32.37	2.00	9.524	0.011	0.005	9.48		

Table B-1. (continued).

Table B-1. (continued).												
Feed coal				Drain		Product coal			Wastewater			
Fresh feed (kg/d)	Wet (kg/d)	H ₂ O (kg/d)	Wet (kg/d)	H ₂ O (kg/d)	Wet (kg/d)	Moisture (%)	H ₂ O (kg/d)	Total (kg/d)	Volume settled solids/total volume	Mass fraction	H ₂ O (kg/d)	
Day												
65	13.31	4.23	0.12	0.402	0.14	7.864	32.55	2.56	1.445	—	0.005	1.44
66	14.96	4.23	0.12	0.436	0.15	3.739	40.17	1.50	10.062	0.010	0.005	10.01
67	23.43	4.23	0.12	0.69	0.24	4.978	36.19	1.80	8.276	0.010	0.005	8.24
68	23.79	4.23	0.12	0.362	0.13	4.86	33.21	1.61	10.024	0.010	0.005	9.98
69	15.57	4.23	0.12	0.362	0.13	8.59	34.93	3.00	10.193	0.005	0.002	10.17
70	18.36	4.23	0.12	0.404	0.14	6.597	39.52	2.61	10.411	0.010	0.005	10.36
71	22.00	4.23	0.12	0.448	0.16	4.835	42.3	2.05	10.471	0.010	0.005	10.42
72	17.65	4.23	0.12	0.44	0.15	8.058	46.57	3.75	10.527	0.020	0.009	10.43
73	20.96	4.23	0.12	0.406	0.14	6.535	44	2.88	10.618	0.008	0.004	10.58
74	19.65	4.23	0.12	0.452	0.16	6.451	44	2.84	10.65	0.005	0.002	10.62
75	21.13	4.23	0.12	0.47	0.16	6.136	44	2.70	10.588	0.005	0.002	10.56
76	15.69	4.23	0.12	0.412	0.14	3.086	46.33	1.43	2.710	0.055	0.026	2.64
77	15.92	4.23	0.12	0.392	0.14	2.768	45.58	1.26	5.585	0.064	0.030	5.42
78	14.46	4.23	0.12	0	0.00	4.945	40.48	2.00	5.188	0.011	0.005	5.16
79	—	4.23	0.12	— ^a	— ^a	— ^a	— ^a	— ^a	—	—	—	—
80	—	4.23	0.12	— ^a	— ^a	— ^a	— ^a	— ^a	—	—	—	—
81	15.70	4.23	0.12	0.142	0.05	6.826	37.74	2.58	5.311	0.005	0.002	5.30
82	13.95	4.23	0.12	0.466	0.16	6.418	43.27	2.78	5.419	0.006	0.003	5.40

Table B-1. (continued).

Table D-1. (Continued).

Day	Fresh feed (kg/d)	Feed coal			Drain		Product coal			Wastewater			
		Wet (kg/d)	H ₂ O (kg/d)		Wet (kg/d)	H ₂ O (kg/d)	Wet (kg/d)	Moisture (%)	H ₂ O (kg/d)	Total (kg/d)	Volume settled solids/total volume	Mass fraction	H ₂ O (kg/d)
83	18.31	4.23	0.12		0.454	0.16	3.850	41.33	1.59	2.918	0.005	0.002	2.91
84	22.08	4.23	0.12										
85	18.51	4.23	0.12		0.462	0.16	5.638	44	2.48	5.220	0.005	0.003	5.21
86	21.20	4.23	0.12		0.508	0.18	4.979	40.79	2.03	5.332	0.006	0.003	5.32
87	19.43	4.23	0.12		0.566	0.20	4.350	44	1.91	5.324	0.005	0.003	5.31
88	13.45	4.23	0.12		0.286	0.10	6.969	42.37	2.95	5.351	0.005	0.002	5.34
89	13.95	4.23	0.12		0.452	0.16	6.848	44	3.01	5.251	0.005	0.003	5.24
90	16.76	4.23	0.12		0.62	0.22	5.380	42.26	2.27	5.302	0.006	0.003	5.29
91	16.71	4.5	0.13		0.446	0.16	6.525	38.96	2.54	5.166	0.005	0.002	5.15
92	15.80	4.5	0.13		0.412	0.14	5.637	44.53	2.51	5.437	0.005	0.002	5.42
93	12.96	4.5	0.13		0.508	0.18	6.707	44	2.95	5.313		0.003	5.30
94	16.03	4.5	0.13		0.456	0.16	6.855	44	3.02	5.303		0.003	5.29
95	10.89	4.5	0.13		0.456	0.16	7.430	44	3.27	5.331	0.006	0.003	5.32
96	12.30	4.5	0.13		0.404	0.14	7.119	39.41	2.81	5.292	0.006	0.003	5.28
97	12.85	4.5	0.13		0.464	0.16	6.872	42.23	2.90	5.243	0.005	0.003	5.23
98	13.01	4.5	0.13		0.41	0.14	6.593	41.72	2.75	5.226	0.005	0.003	5.21
99	13.27	4.5	0.13		0.384	0.13	6.858	41.62	2.85	5.209	0.005	0.002	5.20
100	11.85	4.5	0.13		0.246	0.09	8.075	38.98	3.15	5.178	0.005	0.002	5.17

Table B-1. (continued).

Table B-1. (continued).													
Day	Fresh feed (kg/d)	Feed coal			Drain		Product coal			Wastewater			
		Wet (kg/d)	H ₂ O (kg/d)		Wet (kg/d)	H ₂ O (kg/d)	Wet (kg/d)	Moisture (%)	H ₂ O (kg/d)	Total (kg/d)	Volume settled solids/total volume	Mass fraction	H ₂ O (kg/d)
101	11.09	4.5	0.13		0.346	0.12	7.306	42.81	3.13	5.189	0.010	0.005	5.16
102	12.81	4.5	0.13		0.294	0.10	7.011	44.3	3.11	5.234	0.005	0.002	5.22
103	17.08	4.5	0.13		0.408	0.14	2.872	44	1.26	5.189	—	0.003	5.18
104	12.47	4.5	0.13		0.456	0.16	8.029	42.59	3.42	5.134	0.005	0.002	5.12
105	13.27	4.5	0.13		0	—	1.640	44	—	0.000	—	0.003	—
106	10.80	4.5	0.13		0	0.00	7.306	44	3.21	1.912	0.005	0.002	1.91
107	14.88	4.5	0.13		0	0.00	8.033	33.12	2.66	4.929	—	0.003	4.92
108	9.56	9.84	0.29		0.684	0.24	9.732	42.67	4.15	0.000	—	—	—
109	6.15	9.84	0.29		0.648	0.23	7.868	36.73	2.89	0.000	—	—	—
110	9.60	9.84	0.29		0.784	0.27	7.651	46.83	3.58	0.000	—	—	—
111	10.20	9.84	0.29		0.638	0.22	8.960	34.77	3.12	0.000	—	—	—
112	10.24	9.84	0.29		0.79	0.28	9.580	38.3	3.67	0.000	—	—	—
113	8.75	9.84	0.29		0.62	0.22	11.488	28.15	3.23	0.000	—	—	—
114	8.83	9.84	0.29		0.794	0.28	11.138	36.88	4.11	0.000	—	—	—
115	10.44	9.84	0.29		0.612	0.21	11.614	44	5.11	0.000	—	—	—
116	9.70	9.84	0.29		0.402	0.14	13.534	44	5.95	0.000	—	—	—
117	17.72	9.84	0.29		0.644	0.23	14.949	33.46	5.00	0.000	—	—	—
118	13.27	9.84	0.29		0.664	0.23	13.593	32.4	4.40	0.000	—	—	—

Table B-1. (continued).

TABLE D-7. (Continued).												
Feed coal				Drain		Product coal			Wastewater			
Day	Fresh feed (kg/d)	Wet (kg/d)	H ₂ O (kg/d)	Wet (kg/d)	H ₂ O (kg/d)	Wet (kg/d)	Moisture (%)	H ₂ O (kg/d)	Total (kg/d)	Volume settled solids/		
										total volume	Mass fraction	H ₂ O (kg/d)
119	12.53	9.84	0.29	0.452	0.16	16.094	19.9	3.20	0.000	—	—	—
120	13.35	9.84	0.29	0.584	0.20	16.632	18.79	3.13	0.000	—	—	—
121	12.36	9.84	0.29	0.762	0.27	13.941	18.44	2.57	0.000	—	—	—
122	9.28	9.84	0.29	0.756	0.26	12.190	41.81	5.10	0.000	—	—	—
123	10.13	9.84	0.29	0.586	0.21	11.566	43.9	5.08	0.000	—	—	—
124	10.04	9.84	0.29	0.55	0.19	12.268	45.73	5.61	0.000	—	—	—
125	7.72	9.84	0.29	0.676	0.24	15.432	46.97	7.25	0.000	—	—	—
126	16.38	9.84	0.29	0.498	0.17	14.162	45.78	6.48	0.000	—	—	—
127	12.71	9.84	0.29	0.528	0.18	10.793	42.86	4.63	0.000	—	—	—
128	12.68	9.84	0.29	0.636	0.22	15.754	43.23	6.81	0.000	—	—	—
129	12.20	9.84	0.29	0.576	0.20	13.492	43.12	5.82	0.000	—	—	—
130	11.89	9.84	0.29	0.59	0.21	14.090	42.33	5.96	0.000	—	—	—
131	8.77	9.84	0.29	0.628	0.22	11.628	41.83	4.86	0.000	—	—	—
132	24.54	16.4	0.48	0.518	0.18	25.409	41.75	10.61	0.000	—	—	—
133	18.33	16.4	0.48	0.448	0.16	27.587	40.83	11.26	0.000	—	—	—
134	16.32	16.4	0.48	0.604	0.21	25.595	42.11	10.78	0.000	—	—	—
135	17.88	16.4	0.48	0.548	0.19	25.721	45.3	11.65	0.000	—	—	—
136	13.74	16.4	0.48	0.89	0.31	11.272	53.33	6.01	0.000	—	—	—

Table B-1. (continued).

Day	Feed coal				Drain		Product coal			Wastewater		
	Fresh feed (kg/d)	Wet (kg/d)	H ₂ O (kg/d)	Wet (kg/d)	H ₂ O (kg/d)	Wet (kg/d)	Moisture (%)	H ₂ O (kg/d)	Total (kg/d)	Volume settled solids/total volume	Mass fraction	H ₂ O (kg/d)
136	13.74	16.4	0.48	0.89	0.31	11.272	53.33	6.01	0.000	—	—	—
137	20.14	16.4	0.48	1.462	0.51	16.240	42.47	6.90	0.000	—	—	—
138	16.71	16.4	0.48	1.156	0.40	26.958	43.51	11.73	0.000	—	—	—
139	25.91	16.4	0.48	1.274	0.45	14.423	44.68	6.44	0.000	—	—	—
140	13.12	16.4	0.48	0.886	0.31	33.924	44.4	15.06	0.000	—	—	—
141	18.55	16.4	0.48	1.174	0.41	18.524	43.44	8.05	0.000	—	—	—
142	23.64	16.4	0.48	1.016	0.36	21.493	43.72	9.40	0.000	—	—	—
143	20.84	16.4	0.48	1.168	0.41	24.312	42.86	10.42	0.000	—	—	—
144	16.99	16.4	0.48	1.166	0.41	22.792	43.22	9.85	0.000	—	—	—
145	18.73	16.4	0.48	1.3	0.46	23.459	43.71	10.25	0.000	—	—	—
146	19.88	16.4	0.48	1.394	0.49	21.046	42.17	8.88	0.000	—	—	—
147	16.18	16.4	0.48	1.466	0.51	23.783	42.5	10.11	0.000	—	—	—
148	9.69	16.4	0.48	1.208	0.42	25.999	42.89	11.15	0.000	—	—	—

a. No data available because taking trough effluent and evaporation measurements.

Table B-2. The solids balance.

Day	Feed coal			Drain		Product coal			Wastewater			
	Wet (kg/d)	Dry (kg/d)	Wet (kg/d)	Dry (kg/d)	Wet (kg/d)	Moisture (%)	Dry (kg/d)	Total (kg/d)	Volume settled solids/ total volume	Mass fraction	Solids (kg/d)	
30	4.23	4.11	0.68	0.442	3.925	44	2.198	8.278	0.0104	0.0049	0.041	
31	4.23	4.11	0.09	0.0585	3.287	44	1.841	6.603	—	0.0050	0.033	
32	4.23	4.11	0.09	0.0585	4.18	44	2.341	8.632	0.0120	0.0057	0.049	
33	4.23	4.11	0.09	0.0585	4.154	44	2.326	8.07	0.0084	0.0040	0.032	
34	4.23	4.11	0.23	0.1495	5.208	44	2.916	8.865	0.0109	0.0051	0.045	
35	4.23	4.11	0.314	0.2041	5.897	44	3.302	8.707	0.0323	0.0152	0.132	
36	4.23	4.11	0.234	0.1521	5.865	44	3.284	9.421	0.0160	0.0075	0.071	
37	4.23	4.11	0.18	0.117	5.803	44	3.250	10.329	0.0100	0.0047	0.049	
38	4.23	4.11	0.216	0.1404	6.495	24.3	4.917	10.01	0.0159	0.0075	0.075	
39	4.23	4.11	0.3	0.195	6.9	26.6	5.065	9.627	0.0110	0.0052	0.050	
40	4.23	4.11	0.318	0.2067	5.47	29.3	3.867	9.154	—	0.0050	0.046	
41	4.23	4.11	0.326	0.2119	7.923	24.9	5.950	9.674	0.0123	0.0058	0.056	
42	4.23	4.11	0.328	0.2132	6.463	34.2	4.253	9.852	0.0125	0.0059	0.058	
43	4.23	4.11	0.534	0.3471	6.444	36.6	4.085	12.968	0.0135	0.0064	0.083	

Table B-2. (continued).

Day	Feed coal			Drain		Product coal			Wastewater			
	Wet (kg/d)	Dry (kg/d)		Wet (kg/d)	Dry (kg/d)	Wet (kg/d)	Moisture (%)	Dry (kg/d)	Total (kg/d)	Volume settled solids/ total volume	Mass fraction	Solids (kg/d)
43	4.23	4.11		0.534	0.3471	6.444	36.6	4.085	12.968	0.0135	0.0064	0.083
44	4.23	4.11		0.492	0.3198	6.292	32.9	4.222	9.407	0.0130	0.0061	0.058
45	4.23	4.11		0.472	0.3068	5.758	36.4	3.662	9.603	0.0116	0.0055	0.053
46	4.23	4.11		0.542	0.3523	4.423	45.6	2.406	9.933	0.0052	0.0025	0.024
47	4.23	4.11		0.442	0.2873	5.388	41.1	3.174	9.42	0.0213	0.0100	0.094
48	4.23	4.11		0.344	0.2236	6.382	40	3.829	8.413	0.0100	0.0047	0.040
49	4.23	4.11		0.52	0.338	6.694	31.2	4.605	8.697	0.0115	0.0054	0.047
50	4.23	4.11		0.568	0.3692	7.408	27.9	5.341	8.787	0.0109	0.0051	0.045
51	4.23	4.11		0.546	0.3549	7.939	31.87	5.409	9.47	0.0109	0.0051	0.049
52	4.23	4.11		0.34	0.221	8.018	44	4.490	9.989	0.0114	0.0054	0.054
53	4.23	4.11		0.334	0.2171	7.909	44	4.429	0	—	0.0050	0.000
54	4.23	4.11		0.468	0.3042	6.846	43.07	3.897	9.658	0.0204	0.0096	0.093
55	4.23	4.11		0.534	0.3471	7.934	30	5.554	9.84	0.0204	0.0096	0.095
56	4.23	4.11		0.384	0.2496	8.288	25.7	6.158	9.602	0.0101	0.0048	0.046

Table B-2. (continued).

Day	Feed coal			Drain		Product coal			Wastewater			
	Wet (kg/d)	Dry (kg/d)		Wet (kg/d)	Dry (kg/d)	Wet (kg/d)	Moisture (%)	Dry (kg/d)	Total (kg/d)	Volume settled solids/ total volume	Mass fraction	Solids (kg/d)
57	4.23	4.11		0.41	0.2665	5.609	27.9	4.044	7.636	0.0824	0.0385	0.294
58	4.23	4.11		0.542	0.3523	6.932	44	3.882	9.738	0.0260	0.0122	0.119
59	4.23	4.11		0.31	0.2015	6.232	44	3.490	9.252	0.0111	0.0052	0.049
60	4.23	4.11		0.36	0.234	7.762	44	4.347	9.292	0.0120	0.0057	0.053
61	4.23	4.11		0.402	0.2613	7.111	29.45	5.017	9.395	0.0111	0.0052	0.049
62	4.23	4.11		0.6	0.39	6.883	34	4.543	9.379	0.0729	0.0341	0.320
63	4.23	4.11		0.252	0.1638	7.123	36.7	4.509	9.545	0.0052	0.0024	0.023
64	4.23	4.11		0.35	0.2275	6.165	32.37	4.169	9.524	0.0109	0.0051	0.049
65	4.23	4.11		0.402	0.2613	7.864	32.55	5.304	1.445	—	0.0050	0.007
66	4.23	4.11		0.436	0.2834	3.739	40.17	2.237	10.062	0.0103	0.0049	0.049
67	4.23	4.11		0.69	0.4485	4.978	36.19	3.176	8.276	0.0100	0.0047	0.039
68	4.23	4.11		0.362	0.2353	4.86	33.21	3.246	10.024	0.0100	0.0047	0.047
69	4.23	4.11		0.362	0.2353	8.59	34.93	5.590	10.193	0.0051	0.0024	0.025
70	4.23	4.11		0.404	0.2626	6.597	39.52	3.990	10.411	0.0104	0.0049	0.051

Table B-2. (continued).

Day	Feed coal			Drain		Product coal			Wastewater			
	Wet (kg/d)	Dry (kg/d)		Wet (kg/d)	Dry (kg/d)	Wet (kg/d)	Moisture (%)	Dry (kg/d)	Total (kg/d)	Volume settled solids/ total volume	Mass fraction	Solids (kg/d)
71	4.23	4.11		0.448	0.2912	4.835	42.3	2.790	10.471	0.0101	0.0048	0.050
72	4.23	4.11		0.44	0.286	8.058	46.57	4.305	10.527	0.0200	0.0094	0.099
73	4.23	4.11		0.406	0.2639	6.535	44	3.660	10.618	0.0077	0.0036	0.039
74	4.23	4.11		0.452	0.2938	6.451	44	3.613	10.65	0.0053	0.0025	0.026
75	4.23	4.11		0.47	0.3055	6.136	44	3.436	10.588	0.0051	0.0024	0.026
76	4.23	4.11		0.412	0.2678	3.086	46.33	1.656	2.710	0.0549	0.0258	0.070
77	4.23	4.11		0.392	0.2548	2.768	45.58	1.506	5.585	0.0638	0.0299	0.167
78	4.23	4.11		0	0	4.945	40.48	2.943	5.188	0.0108	0.0051	0.026
79	4.23	4.11		— ^a	— ^a	— ^a	— ^a	— ^a	— ^a	— ^a	— ^a	— ^a
80	4.23	4.11		— ^a	— ^a	— ^a	— ^a	— ^a	— ^a	— ^a	— ^a	— ^a
81	4.23	4.11		0.142	0.0923	6.826	37.74	4.250	5.311	0.0053	0.0025	0.013
82	4.23	4.11		0.466	0.3029	6.418	43.27	3.641	5.419	0.0060	0.0028	0.015
83	4.23	4.11		0.454	0.2951	3.850	41.33	2.259	2.918	0.0053	0.0025	0.007
84	4.23	4.11		— ^a	— ^a	— ^a	— ^a	— ^a	— ^a	— ^a	— ^a	— ^a

Table B-2. (continued).

Day	Feed coal			Drain		Product coal			Wastewater			
	Wet (kg/d)	Dry (kg/d)		Wet (kg/d)	Dry (kg/d)	Wet (kg/d)	Moisture (%)	Dry (kg/d)	Total (kg/d)	Volume settled solids/ total volume	Mass fraction	Solids (kg/d)
84	4.23	4.11	— ^a	— ^a	— ^a	— ^a	— ^a	— ^a	— ^a	— ^a	— ^a	— ^a
85	4.23	4.11	0.462	0.3003		5.638	44	3.158	5.220	0.0054	0.0026	0.013
86	4.23	4.11	0.508	0.3302		4.979	40.79	2.948	5.332	0.0064	0.0030	0.016
87	4.23	4.11	0.566	0.3679		4.350	44	2.436	5.324	0.0053	0.0025	0.013
88	4.23	4.11	0.286	0.1859		6.969	42.37	4.016	5.351	0.0050	0.0024	0.013
89	4.23	4.11	0.452	0.2938		6.848	44	3.835	5.251	0.0053	0.0025	0.013
90	4.23	4.11	0.62	0.403		5.380	42.26	3.106	5.302	0.0065	0.0031	0.016
91	4.5	4.34	0.446	0.2899		6.525	38.96	3.983	5.166	0.0051	0.0024	0.012
92	4.5	4.34	0.412	0.2678		5.637	44.53	3.127	5.437	0.0052	0.0025	0.013
93	4.5	4.34	0.508	0.3302		6.707	44	3.756	5.313	—	0.0025	0.013
94	4.5	4.34	0.456	0.2964		6.855	44	3.839	5.303	—	0.0025	0.013
95	4.5	4.34	0.456	0.2964		7.430	44	4.161	5.331	0.0056	0.0026	0.014
96	4.5	4.34	0.404	0.2626		7.119	39.41	4.314	5.292	0.0060	0.0028	0.015
97	4.5	4.34	0.464	0.3016		6.872	42.23	3.970	5.243	0.0054	0.0025	0.013

Table B-2. (continued).

Day	Feed coal			Drain		Product coal			Wastewater			
	Wet (kg/d)	Dry (kg/d)		Wet (kg/d)	Dry (kg/d)	Wet (t/g/d)	Moisture (%)	Dry (kg/d)	Total (kg/d)	Volume settled solids/ total volume	Mass fraction	Solids (kg/d)
97	4.5	4.34		0.464	0.3016	5.872	42.23	3.970	5.243	0.0054	0.0025	0.013
98	4.5	4.34		0.41	0.2665	6.593	41.72	3.842	5.226	0.0053	0.0025	0.013
99	4.5	4.34		0.384	0.2496	6.858	41.62	4.004	5.209	0.0051	0.0024	0.013
100	4.5	4.34		0.246	0.1599	8.075	38.98	4.928	5.178	0.0052	0.0024	0.013
101	4.5	4.34		0.346	0.2249	7.306	42.81	4.178	5.189	0.0102	0.0048	0.025
102	4.5	4.34		0.294	0.1911	7.011	44.3	3.905	5.234	0.0050	0.0024	0.012
103	4.5	4.34		0.408	0.2652	2.872	44	1.608	5.189	—	0.0025	0.013
104	4.5	4.34		0.456	0.2964	8.029	42.59	4.609	5.134	0.0050	0.0024	0.012
105	4.5	4.34		0	—	1.640	44	—	0.000	—	0.0025	—
106	4.5	4.34		0	0	7.306	44	4.091	1.912	0.0051	0.0024	0.005
107	4.5	4.34		0	0	8.033	33.12	5.372	4.929	—	0.0025	0.012
108	9.84	9.55		0.684	0.4446	9.732	42.67	5.579	0.000	—	—	—
109	9.84	9.55		0.648	0.4212	7.868	36.73	4.978	0.000	—	—	—
110	9.84	9.55		0.784	0.5096	7.651	46.83	4.068	0.000	—	—	—

Table B-2. (continued).

Table B-2. (Continued).												
Day	Feed coal		Drain		Product coal			Wastewater				
	Wet (kg/d)	Dry (kg/d)	Wet (kg/d)	Dry (kg/d)	Wet (kg/d)	Moisture (%)	Dry (kg/d)	Total (kg/d)	Volume settled solids/ total volume	Mass fraction	Solids (kg/d)	
110	9.84	9.55	0.784	0.5096	7.651	46.83	4.068	0.000	—	—	—	
111	9.84	9.55	0.638	0.4147	8.960	34.77	5.845	0.000	—	—	—	
112	9.84	9.55	0.79	0.5135	9.580	38.3	5.911	0.000	—	—	—	
113	9.84	9.55	0.62	0.403	11.488	28.15	8.254	0.000	—	—	—	
114	9.84	9.55	0.794	0.5161	11.138	36.88	7.030	0.000	—	—	—	
115	9.84	9.55	0.612	0.3978	11.614	44	6.504	0.000	—	—	—	
116	9.84	9.55	0.402	0.2613	13.534	44	7.579	0.000	—	—	—	
117	9.84	9.55	0.644	0.4186	14.949	33.46	9.947	0.000	—	—	—	
118	9.84	9.55	0.664	0.4316	13.593	32.4	9.189	0.000	—	—	—	
119	9.84	9.55	0.452	0.2938	16.094	19.9	12.891	0.000	—	—	—	
120	9.84	9.55	0.584	0.3796	16.632	18.79	13.507	0.000	—	—	—	
121	9.84	9.55	0.762	0.4953	13.941	18.44	11.370	0.000	—	—	—	
122	9.84	9.55	0.756	0.4914	12.190	41.81	7.093	0.000	—	—	—	
123	9.84	9.55	0.586	0.3809	11.566	43.9	6.489	0.000	—	—	—	

Table B-2. (continued).

Day	Feed coal		Drain		Product coal			Wastewater			
	Wet (kg/d)	Dry (kg/d)	Wet (kg/d)	Dry (kg/d)	Wet (kg/d)	Moisture (%)	Dry (kg/d)	Total (kg/d)	Volume settled solids/ total volume	Mass fraction	Solids (kg/d)
123	9.84	9.55	0.586	0.3809	11.566	43.9	6.489	0.000	—	—	—
124	9.84	9.55	0.55	0.3575	12.268	45.73	6.658	0.000	—	—	—
125	9.84	9.55	0.676	0.4394	15.432	46.97	8.183	0.000	—	—	—
126	9.84	9.55	0.498	0.3237	14.162	45.78	7.679	0.000	—	—	—
127	9.84	9.55	0.528	0.3432	10.793	42.86	6.167	0.000	—	—	—
128	9.84	9.55	0.636	0.4134	15.754	43.23	8.943	0.000	—	—	—
129	9.84	9.55	0.576	0.3744	13.492	43.12	7.674	0.000	—	—	—
130	9.84	9.55	0.59	0.3835	14.090	42.33	8.125	0.000	—	—	—
131	9.84	9.55	0.628	0.4082	11.628	41.83	6.764	0.000	—	—	—
132	16.4	15.92	0.518	0.3367	25.409	41.75	14.801	0.000	—	—	—
133	16.4	15.92	0.448	0.2912	27.587	40.83	16.323	0.000	—	—	—
134	16.4	15.92	0.604	0.3926	25.595	42.11	14.817	0.000	—	—	—
135	16.4	15.92	0.548	0.3562	25.721	45.3	14.069	0.000	—	—	—
136	16.4	15.92	0.89	0.5785	11.272	53.33	5.261	0.000	—	—	—

Table B-2. (continued).

Day	Feed coal		Drain		Product coal			Wastewater			
	Wet (kg/d)	Dry (kg/d)	Wet (kg/d)	Dry (kg/d)	Wet (kg/d)	Moisture (%)	Dry (kg/d)	Total (kg/d)	Volume settled solids/ total volume	Mass fraction	Solids (kg/d)
136	16.4	15.92	0.89	0.5785	11.272	53.33	5.261	0.000	—	—	—
137	16.4	15.92	1.462	0.9503	16.240	42.47	9.343	0.000	—	—	—
138	16.4	15.92	1.156	0.7514	26.958	43.51	15.228	0.000	—	—	—
139	16.4	15.92	1.274	0.8281	14.423	44.68	7.979	0.000	—	—	—
140	16.4	15.92	0.886	0.5759	33.924	44.4	18.862	0.000	—	—	—
141	16.4	15.92	1.174	0.7631	18.524	43.44	10.477	0.000	—	—	—
142	16.4	15.92	1.016	0.6604	21.493	43.72	12.096	0.000	—	—	—
143	16.4	15.92	1.168	0.7592	24.312	42.86	13.892	0.000	—	—	—
144	16.4	15.92	1.166	0.7579	22.792	43.22	12.941	0.000	—	—	—
145	16.4	15.92	1.3	0.845	23.459	43.71	13.205	0.000	—	—	—
146	16.4	15.92	1.394	0.9061	21.046	42.17	12.171	0.000	—	—	—
147	16.4	15.92	1.466	0.9529	23.783	42.5	13.675	0.000	—	—	—
148	16.4	15.92	1.208	0.7852	25.999	42.89	14.848	0.000	—	—	—

a. No data available because taking trough effluent and evaporation measurements.

a. No data available because taking trough effluent and evaporation measurements.

Table B-3. The ash balance.

Day	Drain			Feed coal			Product coal			
	%Ash	Wet (kg/d)	Ash (kg/d)	Wet (kg/d)	Ash (kg/d)	%Ash	Wet (kg/d)	Moisture (%)	Dry (kg/d)	Ash (kg/d)
31	17.7	0.09	0.010	4.23	0.387	—	—	—	—	—
32	7.5	0.09	0.004	4.23	0.387	9.6	4.18	44.0	2.341	0.224
33	11.3	0.09	0.007	4.23	0.387	—	—	—	—	—
34	15.2	0.23	0.023	4.23	0.387	—	—	—	—	—
35	19.3	0.314	0.039	4.23	0.387	—	—	—	—	—
36	23.4	0.234	0.036	4.23	0.387	9.0	5.865	44.0	3.284	0.294
37	28.7	0.18	0.034	4.23	0.387	—	—	—	—	—
38	26.2	0.216	0.037	4.23	0.387	—	—	—	—	—
39	18.0	0.3	0.035	4.23	0.387	—	—	—	—	—
40	18.1	0.318	0.037	4.23	0.387	—	—	—	—	—
41	18.7	0.326	0.040	4.23	0.387	—	—	—	—	—
42	18.0	0.328	0.038	4.23	0.387	—	—	—	—	—
43	13.1	0.534	0.046	4.23	0.387	8.4	6.444	36.6	4.085	0.343
44	10.4	0.492	0.033	4.23	0.387	—	—	—	—	—
45	11.4	0.472	0.035	4.23	0.387	—	—	—	—	—

Table B-3. (continued).

Day	Drain			Feed coal		Product coal				
	% Ash	Wet (kg/d)	Ash (kg/d)	Wet (kg/d)	Ash (kg/d)	% Ash	Wet (kg/d)	Moisture (%)	Dry (kg/d)	Ash (kg/d)
45	11.4	0.472	0.035	4.23	0.387	—	—	—	—	—
46	13.8	0.542	0.049	4.23	0.387	—	—	—	—	—
47	14.7	0.442	0.042	4.23	0.387	7.9	5.388	41.1	3.174	0.251
48	20.7	0.344	0.046	4.23	0.387	—	—	—	—	—
49	22.3	0.52	0.075	4.23	0.387	—	—	—	—	—
50	15.9	0.568	0.059	4.23	0.387	—	—	—	—	—
51	11.5	0.546	0.041	4.23	0.387	8.1	7.939	31.87	5.409	0.439
52	14.7	0.34	0.033	4.23	0.387	—	—	—	—	—
53	14.0	0.334	0.030	4.23	0.387	—	—	—	—	—
54	10.9	0.468	0.033	4.23	0.387	—	—	—	—	—
55	9.5	0.534	0.033	4.23	0.387	5.4	7.934	30.0	5.554	0.298
56	8.5	0.384	0.021	4.23	0.387	—	—	—	—	—
57	8.5	0.41	0.023	4.23	0.387	—	—	—	—	—
58	9.3	0.542	0.033	4.23	0.387	—	—	—	—	—

Table B-3. (continued).

Day	Drain			Feed coal		Product coal				
	%Ash	Wet (kg/d)	Ash (kg/d)	Wet (kg/d)	Ash (kg/d)	%Ash	Wet (kg/d)	Moisture (%)	Dry (kg/d)	Ash (kg/d)
59	10.4	0.31	0.021	4.23	0.387	—	—	—	—	—
60	12.2	0.36	0.028	4.23	0.387	—	—	—	—	—
61	11.8	0.402	0.031	4.23	0.387	—	—	—	—	—
62	30.8	0.6	0.120	4.23	0.387	10.1	6.883	34.0	4.543	0.459
63	18.5	0.252	0.030	4.23	0.387	—	—	—	—	—
64	15.0	0.35	0.034	4.23	0.387	—	—	—	—	—
65	11.9	0.402	0.031	4.23	0.387	—	—	—	—	—
66	17.0	0.436	0.048	4.23	0.387	—	—	—	—	—
67	11.8	0.69	0.053	4.23	0.387	—	—	—	—	—
68	14.0	0.362	0.033	4.23	0.387	—	—	—	—	—
69	9.9	0.362	0.023	4.23	0.387	5.8	8.59	34.93	5.590	0.325
70	7.5	0.404	0.020	4.23	0.387	6.0	6.597	39.52	3.990	0.238
71	9.2	0.448	0.027	4.23	0.387	—	—	—	—	—
72	8.5	0.44	0.024	4.23	0.387	6.1	8.058	46.57	4.305	0.263

Table B-3. (continued).

Day	Drain			Feed coal			Product coal			
	% Ash	Wet, (kg/d)	Ash (kg/d)	Wet (kg/d)	Ash (kg/d)	% Ash	Wet (kg/d)	Moisture (%)	Dry (kg/d)	Ash (kg/d)
73	11.5	0.406	0.030	4.23	0.387	6.5	6.535	44.0	3.660	0.238
74	9.8	0.452	0.029	4.23	0.387	5.9	6.451	44.0	3.613	0.211
75	9.2	0.47	0.028	4.23	0.387	5.9	6.136	44.0	3.436	0.203
76	11.9	0.412	0.032	4.23	0.387	—	—	—	—	—
77	15.1	0.392	0.038	4.23	0.387	—	—	—	—	—
78	—	0	—	4.23	0.387	—	—	—	—	—
79	—	—	—	4.23	0.387	—	—	—	—	—
80	—	—	—	4.23	0.387	—	—	—	—	—
81	14.1	0.142	0.013	4.23	0.387	—	—	—	—	—
82	14.0	0.466	0.042	4.23	0.387	6.4	6.418	43.27	3.641	0.232
83	15.7	0.454	0.046	4.23	0.387	—	—	—	—	—
84	—	—	—	4.23	0.387	—	—	—	—	—
85	17.6	0.462	0.053	4.23	0.387	—	—	—	—	—
86	18.7	0.508	0.062	4.23	0.387	—	—	—	—	—

Table B-3. (continued).

Day	Drain			Feed coal			Product coal			
	% Ash	Wet (kg/d)	Ash (kg/d)	Wet (kg/d)	Ash (kg/d)	% Ash	Wet (kg/d)	Moisture (%)	Dry (kg/d)	Ash (kg/d)
87	10.3	0.566	0.038	4.23	0.387	—	—	—	—	—
88	18.3	0.286	0.034	4.23	0.387	—	—	—	—	—
89	10.3	0.452	0.030	4.23	0.387	6.1	6.848	44.0	3.835	0.232
90	14.8	0.62	0.060	4.23	0.387	—	—	—	—	—
91	—	0.446	—	4.5	0.411	—	—	—	—	—
92	12.5	0.412	0.033	4.5	0.411	—	—	—	—	—
93	12.2	0.508	0.040	4.5	0.411	—	—	—	—	—
94	14.3	0.456	0.042	4.5	0.411	—	—	—	—	—
95	16.6	0.456	0.049	4.5	0.411	—	—	—	—	—
96	14.8	0.404	0.039	4.5	0.411	—	—	—	—	—
97	14.1	0.464	0.042	4.5	0.411	6.7	6.872	42.23	3.970	0.268
98	14.8	0.41	0.039	4.5	0.411	6.3	6.593	41.72	3.842	0.244
99	16.7	0.384	0.042	4.5	0.411	6.2	6.858	41.62	4.004	0.250
100	16.2	0.246	0.026	4.5	0.411	6.9	8.075	38.98	4.927	0.341

Table B-3. (continued).

Day	Drain			Feed coal			Product coal			
	% Ash	Wet (kg/d)	Ash (kg/d)	Wet (kg/d)	Ash (kg/d)	% Ash	Wet (kg/d)	Moisture (%)	Dry (kg/d)	Ash (kg/d)
101	17.1	0.346	0.039	4.5	0.411	5.9	7.306	42.81	4.178	0.245
102	20.4	0.294	0.039	4.5	0.411	6.4	7.011	44.3	3.905	0.250
103	15.5	0.408	0.041	4.5	0.411	—	—	—	—	—
104	—	0.456	—	4.5	0.411	—	—	—	—	—
105	—	0	—	4.5	0.411	—	—	—	—	—
106	—	0	—	4.5	0.411	—	—	—	—	—
107	12.1	0	—	4.5	0.411	—	—	—	—	—
108	14.6	0.684	0.065	9.84	0.899	—	—	—	—	—
109	21.5	0.648	0.091	9.84	0.899	6.8	7.868	36.73	4.978	0.339
110	16.3	0.784	0.083	9.84	0.899	—	—	—	—	—
111	15.6	0.638	0.065	9.84	0.899	—	—	—	—	—
112	13.5	0.79	0.069	9.84	0.899	—	—	—	—	—
113	17.7	0.62	0.071	9.84	0.899	—	—	—	—	—
114	16.4	0.794	0.085	9.84	0.899	—	—	—	—	—

Table B-3. (continued).

Day	Drain			Feed coal		Product coal				
	c _i Ash	Wet (kg/d)	Ash (kg/d)	Wet (kg/d)	Ash (kg/d)	c _i Ash	Wet (kg/d)	Moisture (%)	Dry (kg/d)	Ash (kg/d)
115	18.9	0.612	0.075	9.84	0.899	—	—	—	—	—
116	18.9	0.402	0.049	9.84	0.899	—	—	—	—	—
117	13.1	0.644	0.055	9.84	0.899	—	—	—	—	—
118	15.1	0.664	0.065	9.84	0.899	6.0	13.593	32.4	9.189	0.550
119	15.8	0.452	0.047	9.84	0.899	—	—	—	—	—
120	17.3	0.584	0.066	9.84	0.899	—	—	—	—	—
121	15.8	0.762	0.078	9.84	0.899	—	—	—	—	—
122	15.6	0.756	0.077	9.84	0.899	—	—	—	—	—
123	17.9	0.586	0.068	9.84	0.899	—	—	—	—	—
124	19.0	0.55	0.068	9.84	0.899	—	—	—	—	—
125	16.3	0.676	0.072	9.84	0.899	6.6	15.432	46.97	8.184	0.540
126	14.0	0.498	0.045	9.84	0.899	5.6	14.162	45.78	7.679	0.432
127	19.2	0.528	0.066	9.84	0.899	6.0	10.793	42.86	6.167	0.368
128	17.9	0.636	0.074	9.84	0.899	5.8	15.754	43.23	8.944	0.521

Table B-3. (continued).

Day	Drain			Feed coal			Product coal			
	% Ash	Wet (kg/d)	Ash (kg/d)	Wet (kg/d)	Ash (kg/d)	% Ash	Wet (kg/d)	Moisture (%)	Dry (kg/d)	Ash (kg/d)
129	16.4	0.576	0.061	9.84	0.899	6.1	13.492	43.12	7.674	0.471
130	14.8	0.59	0.057	9.84	0.899	5.5	14.09	42.33	8.126	0.448
131	15.4	0.628	0.063	9.84	0.899	6.3	11.628	41.83	6.764	0.424
132	18.8	0.518	0.063	16.4	1.499	—	—	—	—	—
133	21.8	0.448	0.063	16.4	1.499	—	—	—	—	—
134	23.8	0.604	0.093	16.4	1.499	—	—	—	—	—
135	26.1	0.548	0.093	16.4	1.499	6.5	25.721	45.3	14.069	0.907
136	19.5	0.89	0.113	16.4	1.499	—	—	—	—	—
137	19.4	1.462	0.184	16.4	1.499	—	—	—	—	—
138	18.6	1.156	0.140	16.4	1.499	—	—	—	—	—
139	16.7	1.274	0.139	16.4	1.499	—	—	—	—	—
140	23.0	0.886	0.132	16.4	1.499	7.0	33.924	44.4	18.862	1.326
141	16.5	1.174	0.126	16.4	1.499	—	—	—	—	—
142	23.2	1.016	0.153	16.4	1.499	—	—	—	—	—

Table B-3. (continued).

Day	Drain			Feed coal		Product coal				
	% Ash	Wet (kg/d)	Ash (kg/d)	Wet (kg/d)	Ash (kg/d)	% Ash	Wet (kg/d)	Moisture (%)	Dry (kg/d)	Ash (kg/d)
143	18.9	1.168	0.144	16.4	1.499	—	—	—	—	—
144	17.8	1.166	0.135	16.4	1.499	6.3	22.792	43.22	12.941	0.810
145	17.0	1.3	0.143	16.4	1.499	6.2	23.459	43.71	13.205	0.820
146	15.5	1.394	0.141	16.4	1.499	6.3	21.046	42.17	12.171	0.761
147	16.6	1.466	0.158	16.4	1.499	—	—	—	—	—
148	16.8	1.208	0.132	16.4	1.499	7.0	25.999	42.89	14.848	1.040

Table B-4. The iron balance.

Day	Feed coal				Product coal				Wastewater				Drain			
	Wet (kg/d)		Pyritic Fe (kg/d)		Wet (kg/d)		Moist (%)		HCl wash g Fe/g wet coal		Soluble Fe (kg/d)		Pyritic Fe (%)		Pyritic Fe (kg/d)	
	Wet (kg/d)		Pyritic Fe (kg/d)		Wet (kg/d)		Moist (%)		HCl wash g Fe/g wet coal		Soluble Fe (kg/d)		Pyritic Fe (%)		Pyritic Fe (kg/d)	
30	4.23	0.036473	3.925	44	0.001236	0.004852	0.147	0.003231	8.24	0.5375	0.004428	0.68	0.442	—	—	—
31	4.23	0.036473	3.287	44	—	—	—	—	6.57	1.2	0.007884	0.09	0.0585	—	—	—
32	4.23	0.036473	4.18	44	0.001452	0.006071	0.231	0.005407	8.58	1.513	0.012986	0.09	0.0585	—	—	—
33	4.23	0.036473	4.154	44	—	—	—	—	8.04	1.588	0.012764	0.09	0.0585	—	—	—
34	4.23	0.036473	5.208	44	—	—	—	—	8.82	1.338	0.011801	0.23	0.1495	—	—	—
35	4.23	0.036473	5.897	44	—	—	—	—	8.57	1.15	0.009861	0.314	0.2041	—	—	—
36	4.23	0.036473	5.865	44	0.001485	0.00871	0.257	0.008441	9.35	1.475	0.013791	0.234	0.1521	—	—	—
37	4.23	0.036473	5.803	44	—	—	—	—	10.28	1.213	0.01247	0.18	0.117	—	—	—
38	4.23	0.036473	6.495	24.3	—	—	—	—	9.94	1.025	0.010183	0.216	0.1404	—	—	—
39	4.23	0.036473	6.9	26.6	—	—	—	—	9.58	0.825	0.007901	0.3	0.195	—	—	—
40	4.23	0.036473	5.47	29.3	—	—	—	—	9.11	0.46	0.00419	0.318	0.2067	—	—	—
41	4.23	0.036473	7.923	24.9	—	—	—	—	9.62	0.515	0.004953	0.326	0.2119	—	—	—
42	4.23	0.036473	6.463	34.2	—	—	—	—	9.79	0.459	0.004495	0.328	0.2132	—	—	—
43	4.23	0.036473	6.444	36.6	0.0014	0.009022	0.253	0.010336	12.89	0.813	0.010476	0.534	0.3471	—	—	—
44	4.23	0.036473	6.292	32.9	—	—	—	—	9.35	1.05	0.009817	0.492	0.3198	—	—	—
45	4.23	0.036473	5.758	36.4	—	—	—	—	9.55	0.663	0.006332	0.472	0.3068	—	—	—
46	4.23	0.036473	4.423	45.6	—	—	—	—	9.91	0.266	0.002636	0.542	0.3523	—	—	—
47	4.23	0.036473	5.388	41.1	0.001007	0.005426	0.254	0.008061	9.33	0.663	0.006183	0.442	0.2873	—	—	—
48	4.23	0.036473	6.382	40	—	—	—	—	8.37	0.325	0.002721	0.344	0.2236	—	—	—
49	4.23	0.036473	6.694	31.2	—	—	—	—	8.65	0.263	0.002275	0.52	0.338	—	—	—
50	4.23	0.036473	7.408	27.9	—	—	—	—	8.74	0.225	0.001967	0.568	0.3692	—	—	—
51	4.23	0.036473	7.939	31.87	0.001962	0.01558	0.234	0.012657	9.42	0.213	0.002007	0.546	0.3549	1.177	0.004177173	—

Table B-4. (continued).

Feed coal				Product coal				Wastewater				Drain				
Day	Wet (kg/d)	Pyritic Fe (kg/d)		Wet (kg/d)	Moist (%)	HCl wash g Fe/g wet coal	Soluble Fe (kg/d)	Pyritic Fe (%)	Pyritic Fe (kg/d)	H ₂ O (kg/d)	Soluble Fe g/L	Soluble Fe (kg/d)	Wet (kg/d)	Dry (kg/d)	Pyritic Fe (%)	Pyritic Fe (kg/d)
51	4.23	0.036473		7.939	31.87	0.001962	0.01558	0.234	0.012657	9.42	0.213	0.002007	0.546	0.3549	1.177	0.004177173
52	4.23	0.036473		8.018	44	—	—	—	—	9.94	0.763	0.007581	0.34	0.221	—	—
53	4.23	0.036473		7.909	44	—	—	—	—	0.00	—	0	0.334	0.2171	—	—
54	4.23	0.036473		6.846	43.07	—	—	—	—	9.57	0.675	0.006456	0.468	0.3042	—	—
55	4.23	0.036473		7.934	30	0.000779	0.006177	0.274	0.015217	9.75	0.588	0.00573	0.534	0.3471	—	—
56	4.23	0.036473		8.288	25.7	—	—	—	—	9.56	0.9	0.008601	0.384	0.2496	—	—
57	4.23	0.036473		5.609	27.9	—	—	—	—	7.34	1.25	0.009177	0.41	0.2665	—	—
58	4.23	0.036473		6.932	44	—	—	—	—	9.62	1.6	0.01539	0.542	0.3523	—	—
59	4.23	0.036473		6.232	44	—	—	—	—	9.20	1.775	0.016336	0.31	0.2015	—	—
60	4.23	0.036473		7.762	44	—	—	—	—	9.24	1.938	0.017906	0.36	0.234	—	—
61	4.23	0.036473		7.111	29.45	—	—	—	—	9.35	1.45	0.013551	0.402	0.2613	—	—
62	4.23	0.036473		6.883	34	0.001696	0.011673	0.473	0.021487	9.06	1.363	0.012347	0.6	0.39	—	—
63	4.23	0.036473		7.123	36.7	—	—	—	—	9.52	1.538	0.014644	0.252	0.1638	—	—
64	4.23	0.036473		6.165	32.37	—	—	—	—	9.48	1.45	0.013739	0.35	0.2275	—	—
65	4.23	0.036473		7.864	32.55	—	—	—	—	1.44	1.413	0.002032	0.402	0.2613	—	—
66	4.23	0.036473		3.739	40.17	—	—	—	—	10.01	2.15	0.021528	0.436	0.2834	—	—
67	4.23	0.036473		4.978	36.19	—	—	—	—	8.24	1.313	0.010815	0.69	0.4485	—	—
68	4.23	0.036473		4.86	33.21	—	—	—	—	9.98	1.138	0.011353	0.362	0.2353	—	—
69	4.23	0.036473		8.59	34.93	0.001214	0.010425	0.19	0.01062	10.17	1.1	0.011185	0.362	0.2353	—	—
70	4.23	0.036473		6.597	39.52	0.001591	0.010495	0.186	0.007421	10.36	1.5	0.01554	0.404	0.2626	—	—
71	4.23	0.036473		4.835	42.3	—	—	—	—	10.42	1.588	0.016549	0.448	0.2912	—	—
72	4.23	0.036473		8.058	46.57	0.001364	0.010994	0.179	0.007707	10.43	2	0.020856	0.44	0.286	—	—

Table B-4. (continued).

Table B-4. (Continued)																
Day	Feed coal			Product coal						Wastewater				Drain		
	Wet (kg/d)	Pyritic Fe (kg/d)		Wet (kg/d)	Moist (%)	HCl wash g Fe/g wet coal	Soluble Fe (kg/d)	Pyritic Fe (%)	Pyritic Fe (kg/d)	H ₂ O (kg/d)	Soluble Fe g/L	Soluble Fe (kg/d)	Wet (kg/d)	Dry (kg/d)	Pyritic Fe (%)	Pyritic Fe (kg/d)
73	4.23	0.036473		6.535	44	0.001273	0.008317	0.183	0.006697	10.58	2.288	0.024206	0.406	0.2639	—	—
74	4.23	0.036473		6.451	44	0.001735	0.011196	0.185	0.006683	10.62	1.6	0.016998	0.452	0.2938	—	—
75	4.23	0.036473		6.136	44	0.00082	0.005033	0.177	0.006082	10.56	1.125	0.011883	0.47	0.3055	—	—
76	4.23	0.036473		3.086	46.33	—	—	—	—	2.64	1.225	0.003234	0.412	0.2678	—	—
77	4.23	0.036473		2.768	45.58	—	—	—	—	5.42	1.588	0.008603	0.392	0.2548	—	—
78	4.23	0.036473		4.945	40.48	—	—	—	—	5.16	1.55	0.008001	0	0	—	—
79	4.23	0.036473		— ^a	— ^a	— ^a	— ^a	— ^a	— ^a	—	—	0	—	—	—	—
80	4.23	0.036473		— ^a	— ^a	— ^a	— ^a	— ^a	— ^a	—	—	0	—	—	—	—
81	4.23	0.036473		6.826	37.74	—	—	—	—	5.30	1.763	0.009341	0.142	0.0923	—	—
82	4.23	0.036473		6.418	43.27	0.001815	0.011649	—	—	5.40	1.875	0.010131	0.466	0.3029	—	—
83	4.23	0.036473		3.850	41.33	—	—	—	—	2.91	2.388	0.00695	0.454	0.2951	—	—
84	4.23	0.036473		— ^a	— ^a	— ^a	— ^a	— ^a	— ^a	—	2.438	0	—	—	—	—
85	4.23	0.036473		5.638	44	—	—	—	—	5.21	1.938	0.010091	0.462	0.3003	—	—
86	4.23	0.036473		4.979	40.79	—	—	—	—	5.32	1.638	0.008707	0.508	0.3302	—	—
87	4.23	0.036473		4.350	44	—	—	—	—	5.31	0.988	0.005247	0.566	0.3679	—	—
88	4.23	0.036473		6.969	42.37	—	—	—	—	5.34	1.325	0.007074	0.286	0.1859	—	—
89	4.23	0.036473		6.848	44	0.001991	0.013636	0.095	0.003643	5.24	0.675	0.003536	0.452	0.2938	—	—
90	4.23	0.036473		5.380	42.26	—	—	—	—	5.29	1.363	0.007204	0.62	0.403	—	—
90	4.23	0.036473		5.7674	—	—	0.00933	0.2212 0	0.012757	8.20	1.22	0.009	—	0.2552	—	—
91	4.5	0.038801		6.525	38.96	—	—	—	—	5.15	1.213	0.006251	0.446	0.2899	—	—
92	4.5	0.038801		5.637	44.53	—	—	—	—	5.42	1.388	0.007527	0.412	0.2678	—	—

Table B-4. (continued).

Table B-4. (continued).																
Day	Feed coal			Product coal						Wastewater			Drain			
	Wet (kg/d)	Pyritic Fe (kg/d)		Wet (kg/d)	Moist (%)	HCl wash g Fe/g wet coal	Soluble Fe (kg/d)	Pyritic Fe (%)	Pyritic Fe (kg/d)	H ₂ O (kg/d)	Soluble Fe g/L	Soluble Fe (kg/d)	Wet (kg/d)	Dry (kg/d)	Pyritic Fe (%)	Pyritic Fe (kg/d)
93	4.5	0.038801	6.707	44	—	—	—	—	—	5.30	1.8	0.009539	0.508	0.3302	—	—
94	4.5	0.038801	6.855	44	—	—	—	—	—	5.29	—	0	0.456	0.2964	—	—
95	4.5	0.038801	7.430	44	—	—	—	—	—	5.32	1.85	0.009836	0.456	0.2964	—	—
96	4.5	0.038801	7.119	39.41	—	—	—	—	—	5.28	1.525	0.008048	0.404	0.2626	—	—
97	4.5	0.038801	6.872	42.23	0.002172	0.014925	0.101	0.00401	0.00401	5.23	0.85	0.004445	0.464	0.3016	3.329	0.010040264
98	4.5	0.038801	6.593	41.72	0.002294	0.015123	0.107	0.004111	0.004111	5.21	1.575	0.00821	0.41	0.2665	1.875	0.007932288
99	4.5	0.038801	6.858	41.62	0.002303	0.015796	0.114	0.004564	0.004564	5.20	1.338	0.006952	0.384	0.2496	3.178	0.007932288
100	4.5	0.038801	8.075	38.98	0.002274	0.018367	0.137	0.006751	0.006751	5.17	2.025	0.010461	0.246	0.1599	3.883	0.006208917
101	4.5	0.038801	7.306	42.81	0.002022	0.014772	0.108	0.004512	0.004512	5.16	1.963	0.010136	0.346	0.2249	3.041	0.006839209
102	4.5	0.038801	7.011	44.3	0.002137	0.014984	0.11	0.004296	0.004296	5.22	2.075	0.010834	0.294	0.1911	2.777	0.005306847
103	4.5	0.038801	2.872	44	—	—	—	—	—	5.18	2.075	0.010741	0.408	0.2652	—	—
104	4.5	0.038801	8.027	42.59	—	—	—	—	—	5.12	1.725	0.008835	0.456	0.2964	—	—
105	4.5	0.038801	1.640	—	—	—	—	—	—	—	—	0	0	—	—	—
106	4.5	0.038801	7.306	44	—	—	—	—	—	1.91	0.325	0.001574	0	0	—	—
107	4.5	0.038801	8.033	33.12	—	—	—	—	—	4.92	0.775	0.003811	0	0	—	—
107	4.50	0.038801	6.522	—	—	0.0157	0.1128	0.007359	0.007359	5.005	1.533	0.0069	—	0.2312	—	—
108	9.84	0.084845	9.732	42.67	—	—	—	—	—	—	2.025	0	0.684	0.4446	—	—
109	9.84	0.084845	7.868	36.73	0.002441	0.019208	0.176	0.008762	0.008762	—	2.2	0	0.648	0.4212	—	—
110	9.84	0.084845	7.651	46.83	—	—	—	—	—	—	2.188	0	0.784	0.5096	—	—
111	9.84	0.084845	8.960	34.77	—	—	—	—	—	—	2.563	0	0.638	0.4147	—	—
112	9.84	0.084845	9.580	38.3	—	—	—	—	—	—	2.438	0	0.79	0.5135	—	—
113	9.84	0.084845	11.488	28.15	—	—	—	—	—	—	3.288	0	0.62	0.403	—	—

Table B-4. (continued).

Table D-4. (Continued).																
Feed coal			Product coal					Wastewater				Drain				
Day	Wet (kg/d)	Pyritic Fe (kg/d)	Wet (kg/d)	Moist (%)	HCl wash g Fe/g wet coal	Soluble Fe (kg/d)	Pyritic Fe (%)	Pyritic Fe (kg/d)	H ₂ O (kg/d)	Soluble Fe g/L	Soluble Fe (kg/d)	Wet (kg/d)	Dry (kg/d)	Pyritic Fe (%)	Pyritic Fe (kg/d)	
114	9.84	0.084845	11.138	36.88	—	—	—	—	—	2.225	0	0.794	0.5161	—	—	
115	9.84	0.084845	11.614	44	—	—	—	—	—	—	0	0.612	0.3978	—	—	
116	9.84	0.084845	13.534	44	—	—	—	—	—	—	0	0.402	0.2613	—	—	
117	9.84	0.084845	14.949	33.46	—	—	—	—	—	1.888	0	0.644	0.4186	—	—	
118	9.84	0.084845	13.593	32.4	0.00264	0.035882	0.203	0.018653	—	2.388	0	0.664	0.4316	—	—	
119	9.84	0.084845	16.094	19.9	—	—	—	—	—	1.388	0	0.452	0.2938	—	—	
120	9.84	0.084845	16.632	18.79	—	—	—	—	—	3.375	0	0.584	0.3796	—	—	
121	9.84	0.084845	13.941	18.44	—	—	—	—	—	4.263	0	0.762	0.4953	—	—	
122	9.84	0.084845	12.190	41.81	—	—	—	—	—	4.3	0	0.756	0.4914	—	—	
123	9.84	0.084845	11.566	43.9	—	—	—	—	—	3.988	0	0.586	0.3809	—	—	
124	9.84	0.084845	12.268	45.73	—	—	—	—	—	5.788	0	0.55	0.3575	—	—	
125	9.84	0.084845	15.432	46.97	0.002739	0.042272	0.213	0.017431	—	—	0	0.676	0.4394	2.509	0.011024546	
126	9.84	0.084845	14.162	45.78	0.003424	0.048492	0.16	0.012286	—	4.4	0	0.498	0.3237	2.374	0.007684638	
127	9.84	0.084845	10.793	42.86	0.002333	0.025184	0.188	0.011594	—	4.488	0	0.528	0.3432	3.922	0.013460304	
128	9.84	0.084845	15.754	43.23	0.004366	0.068778	0.186	0.016635	—	4.425	0	0.636	0.4134	3.494	0.014444196	
129	9.84	0.084845	13.492	43.12	0.003586	0.048379	0.218	0.016729	—	5.513	0	0.576	0.3744	3.041	0.011385504	
130	9.84	0.084845	14.090	42.33	0.003908	0.055062	0.193	0.015682	—	4.088	0	0.59	0.3835	4.327	0.016594045	
131	9.84	0.084845	11.628	41.83	0.003187	0.037053	0.217	0.014678	—	3.213	0	0.628	0.4082	2.529	0.010323378	
131	9.84	0.084845	12.423	—	—	0.04226	0.1948 ₉	0.024211	—	—	0	—	0.4090	—	—	
132	16.4	0.141409	25.409	41.75	—	—	—	—	—	4.138	0	0.518	0.3367	—	—	
133	16.4	0.141409	27.587	40.83	—	—	—	—	—	3.475	0	0.448	0.2912	—	—	

Table B-4. (continued).

Table B-4. (continued).

Feed coal			Product coal						Wastewater			Drain			
Day	Wet (kg/d)	Pyritic Fe (kg/d)	Wet (kg/d)	Moist (%)	HCl wash g Fe/g wet coal	Soluble Fe (kg/d)	Pyritic Fe (%)	Pyritic Fe (kg/d)	H ₂ O (kg/d)	Soluble Fe g/L	Soluble Fe (kg/d)	Wet (kg/d)	Dry (kg/d)	Pyritic Fe (%)	Pyritic Fe (kg/d)
134	16.4	0.141409	25.595	42.11	—	—	—	—	—	1.463	0	0.604	0.3926	—	—
135	16.4	0.141409	25.721	45.3	0.00174	0.044756	0.305	0.042911	—	1.813	0	0.548	0.3562	—	—
136	16.4	0.141409	11.272	53.33	—	—	—	—	—	1.35	0	0.89	0.5785	—	—
137	16.4	0.141409	16.240	42.47	—	—	—	—	—	1.538	0	1.462	0.9503	—	—
138	16.4	0.141409	26.958	43.51	—	—	—	—	—	1.2	0	1.156	0.7514	—	—
139	16.4	0.141409	14.423	44.68	—	—	—	—	—	1.65	0	1.274	0.8281	—	—
140	16.4	0.141409	33.924	44.4	0.001869	0.063403	0.432	0.081483	—	2.9	0	0.886	0.5759	—	—
141	16.4	0.141409	18.524	43.44	—	—	—	—	—	3.188	0	1.174	0.7631	—	—
142	16.4	0.141409	21.493	43.72	—	—	—	—	—	3.15	0	1.016	0.6604	—	—
143	16.4	0.141409	24.312	42.86	—	—	—	—	—	3.025	0	1.168	0.7592	—	—
144	16.4	0.141409	22.792	43.22	0.001661	0.037863	0.271	0.035071	—	2.863	0	1.166	0.7579	—	—
145	16.4	0.141409	23.459	43.71	0.002923	0.068562	0.355	0.046877	—	2.8	0	1.3	0.845	—	—
146	16.4	0.141409	21.046	42.17	0.002782	0.058544	0.333	0.04053	—	2.838	0	1.394	0.9061	—	—
147	16.4	0.141409	23.783	42.5	—	—	—	—	—	2.725	0	1.466	0.9529	—	—
148	16.4	0.141409	25.999	42.89	0.002126	0.055263	0.366	0.054344	—	3.163	0	1.208	0.7852	—	—
148	16.400	0.141409	22.855	—	—	0.0547	0.3436	0.05020	—	—	—	—	0.6759	—	—

a. No data available because taking trough and effluent measurements.

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**DATE
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END