# Chemical Comminution of Coal

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## Introduction

Due to the problems associated with importing oil from abroad and the rapid increase in the price of crude oil over the past decade, coal is becoming a primary alternative fuel of the future. However, the high cost of extraction and increased concern for environmental standards and for the health and safety of the miners limits the production of coal by conventional technologies. As a result there is increased interest in chemical mining methods. These methods promise improved economics and they will expand minable coal reserves.

The term comminution literally means breaking into small particles. Comminution can be carried out by physical means such as grinding or crushing machinery, or the detonation of explosives. It can also be achieved by chemical action. A method devised by Aldrich (1984) for the underground chemical comminution of coal is shown in Figure 1. The process involves drilling two bore holes extending from the surface to separate points in a coal seam. A reactive reagent is pumped through one hole to the seam, where chemical action forms a slurry of coal particles and reagent. The second bore hole permits outflow of the slurry. Surface facilities separate the coal particles from the slurry and the reagent is recycled after treatment.

This type of mining technology may be limited by problems in four areas of concern:

1. Removal of slurry to the surface
2. Heat transfer to the overburden
3. Possible contamination of water reservoirs
4. Reactivity of reagent with the coal

Due to the complexity of the forces acting on coal, it is difficult to propose a definite mechanism for chemical comminution. Nevertheless, researchers have argued a sequence of events that involves diffusion to reactive sites, ionic substitution on polar bonds, swelling of the matrix, and finally, internal breakage (Dryden, 1952; McRee, 1977), Skidmore, (1974).

The objective of the present research is to study the chemical reactivity of a mixture of methyl alcohol and aqueous sodium hydroxide solution in the temperature range 298 to 363 K, and a caustic concentration of 0 to 10 wt. %, on an Iowa bituminous coal.

The sample studied was collected from coal zone 4, equivalent to most historical references to Laddsdale coal (Van Dorpe et al., 1984). The coals in this zone are typical high-sulfur, high-ash middle Pennsylvania Cherokee group coals. The apparent rank is high-volatile C bituminous coal. The relatively high content of sulfur and 23 other elements in these coals is related to near neutral (6–8) pH conditions in the depositional and early diagenetic environments, and to postdepositional sphalerite/calcite/pyrite/kaolinite/barite mineralization. Table 1 shows the typical proximate analysis of the coal studied in this work.

## Experimental Procedures

In a batch system, a series of experiments was conducted by allowing 50 mm cubic pieces of coal to react for 8 h at 308 K with a 2.5 molar solution of sodium hydroxide and methyl alcohol. A low caustic concentration (0–10 wt. %) was used throughout the experiments because a number of scouting experiments with higher concentrations of sodium hydroxide led to poor comminution rates. The vapors were refluxed so that the reagent concentration remained constant during each experiment. When the time of the reaction had elapsed, the coal and reagent were separated by filtering. The comminuted coal particles were rinsed with distilled water and allowed to dry for two days in an oven. Then the morphic characteristics of the particles were analyzed and the comminution rates were determined.

Comminution is defined as the rate of surface area generation. The surface area generated in the experiments was determined by performing sieve analysis on the comminuted coal particles. Seven size ranges were obtained using a machine called a shape analyzer TM. An average particle diameter was taken to be the arithmetic average in each size range. Using these average particle diameters, an approximate surface area per gram of coal in each sieve tray was obtained. The total new surface area

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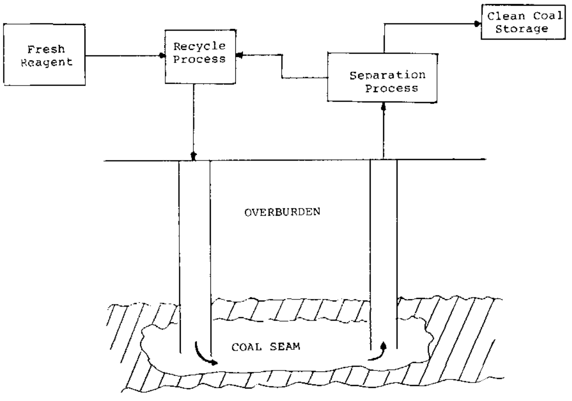

**Figure 1. A concept for *in situ* disintegration of coal.**

was subtracted from the initial surface area of the sample to yield the new surface area generated. These values were used along with the reaction time to compare results for the sets of conditions investigated.

## Comminution Rates

A series of batch preliminary experiments at 298 K was conducted to determine the comminution rates for various reagents and equimolar mixtures of reagents. Table 2 lists the results obtained from this investigation.

The data seem to indicate that one of the many factors that may be controlling the rate of comminution is the molecular size of the reagent. A reagent of small molecular size (e.g., methanol, molecular size = 25.2 Å) gives rise to a higher comminution rate than a reagent of larger molecular size. A possible explanation for this phenomenon is that the smaller molecules diffuse through the porous structure of the coal more easily and thus have more reactive surface area available to them.

The results from sieve analysis revealed that the comminution rate decreased with a fall in the reaction temperature in a caustic-methanol system, Figure 2. Higher temperatures seem to increase the molecular interactions and mobility of the reagent and cause faster and deeper penetration into the coal matrix. Figure 3 is a plot of comminution rate as a function of caustic concentration in the caustic-methanol system. The molar water/alcohol ratio in these systems ranged from 0.0125 to 1.00. Although the data points are somewhat scattered, one can still draw a quantitative conclusion from them. It appears that the higher comminution rates occur between 6 and 8 wt. % caustic.

**Table 1. Typical Proximate Analysis of Experimental Coal**

| | Composition as Received, % |
|---|---|
| Moisture | 19.83 |
| Ash | 9.13 |
| Volatile | 31.92 |
| Fixed carbon | 39.12 |
| Total | 100.00 |
| kJ/kg | $2.38 \times 10^4$ |
| Sulfur, % | 1.91 |

**Table 2. Results of Scouting Experiments**

| Reagent | Comminution Rate mg/cm$^2$ h | Molecular Size, Å |
|---|---|---|
| Methanol | 3.6 | 25.20 |
| Ethanol | 3.1 | 28.50 |
| Propanol | 3.0 | 30.96 |
| Butanol | 2.8 | 33.11 |
| Pentanol | 2.1 | 35.01 |
| Hexanol | 2.1 | 36.79 |
| Acetone | 19.6 | 30.78 |
| NaOH | 3.9 | 19.53 |
| Acetone-NaOH | 16.4 | — |
| Methanol-NaOH | 18.5 | — |
| Ethanol-NaOH | 18.1 | — |
| Propanol-NaOH | 14.3 | — |
| Butanol-NaOH | 12.6 | — |
| Pentanol-NaOH | 12.6 | — |
| Hexanol-NaOH | 10.3 | — |
| Sodium Kraft liquor (K2S+NaOH) | 10.2 | — |
| Lithium Kraft liquor (Li2S+NaOH) | 8.4 | — |

## Morphological Studies

Information from morphological studies of chemically comminuted particles could be of great significance in slurry transport to the surface, as previous studies have shown that the size and shape of the particles play a major role in slurry transport (Vetter, 1983).

The shape of the comminuted coal particles was analyzed by the shape analyzer TM machine. Figures 4 and 5 show the respective effects of caustic concentration and reaction temperature on the not-roundness of the particles. Luerkens et al. (1982) developed a system of morphological descriptors that mathematically define the shape of irregular (nonspherical) particles. This system was used to determine the not-roundness value of the comminuted particles. Not-roundness is the deviation of the particle profile from that of a circle. A perfect circle has a not-

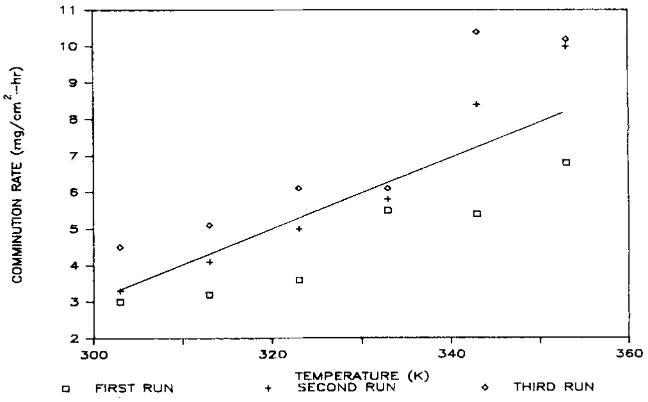

**Figure 2. Comminution rate as a function of reaction temperature in a caustic-methanol system, molar ratio = 0.11.**

Higher temperatures yielded higher rates as diffusivity of reagent into coal matrix increased.

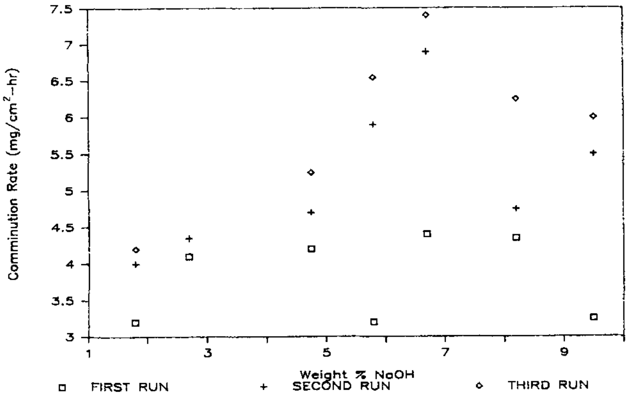

**Figure 3. Comminution rate as a function of caustic concentration in a caustic-methanol system at 308 K.**

Data points are scattered, but maximum rate seems to occur between 6 and 8 wt. % NaOH.

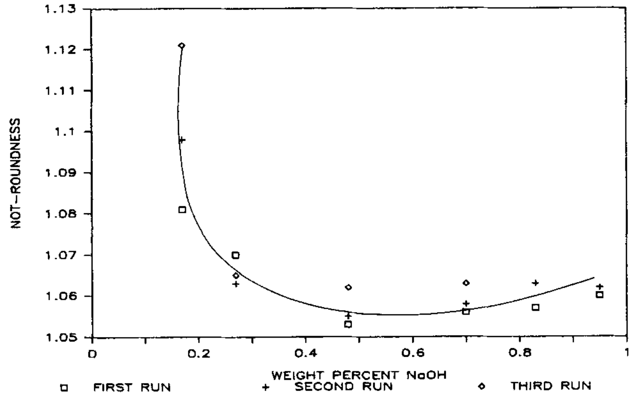

**Figure 4. Not-roundness of comminuted particles as a function of sodium hydroxide concentration.**

Higher concentrations yielded more circular and smoother particles in a caustic-methanol system.

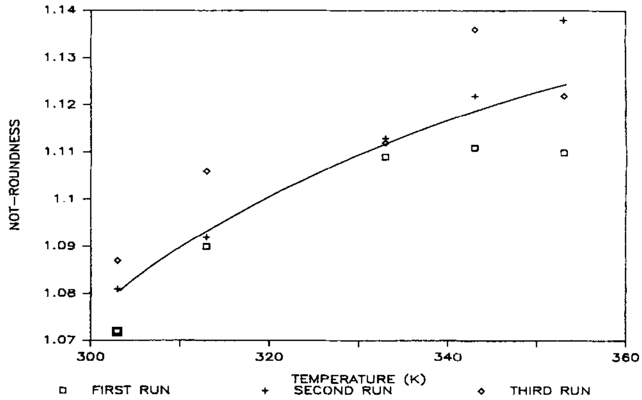

**Figure 5. Not-roundness of comminuted particles as a function of reaction temperature in a caustic-methanol system, molar ratio = 0.11.**

Higher temperatures yielded rougher and less circular particles.

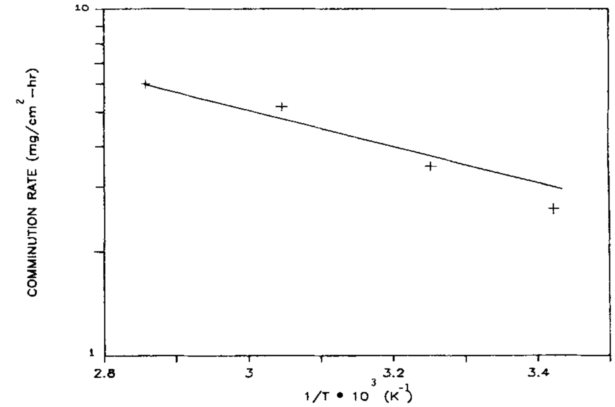

**Figure 6. Comminution rate vs. reciprocal temperature in a caustic-methanol system, molar ratio = 0.11.**

Slope of line represents activation energy of comminution.

roundness value of 1.00; the value is higher than 1.00 for other shapes. Thus a low not-roundness value indicates that a particle is more nearly circular. The values found in this work indicate that higher concentrations of NaOH or lower reaction temperatures make particles smoother and more round.

## Kinetic Studies

Temperature effects were correlated to determine if a consistent mechanism exists for comminution rates. Song (1981) concluded that the rate of reaction is proportional to a power of the concentration of coal and reagent.

For the purposes of this analysis, it was convenient to assume that the weight of coal and the concentration of the reagent remained constant throughout the reaction period. These are valid assumptions, since the specific gravity of the reagent remained constant during the experiment and the coal weight loss was only 2%.

If a consistent mechanism exists, a plot of $\ln(dw/dt)$ vs. $1/T$ would yield a straight line, as shown in Figure 6. From this plot the activation energy was calculated to be 58.9 kJ/gmol. The high magnitude of activation energy indicates that chemical reactions are taking place and may be controlling the rate of comminution.

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