Correlation Method for Chemical Comminution of Coal

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In spite of many experimental studies of the chemical comminution of coal, there have been only a few reported attempts to correlate experimental data and mathematically model the process. Keller and Smith (1976) presented a strain energy model based on the thermodynamic analysis. Civan et al. (1987) proposed the "capillary-imbibition number" as an important parameter for characterization of chemicals used in comminution. Civan and Knapp (1989) developed a phenomenological model for chemical comminution to study the relative effects of the governing processes. Sensitivity studies carried out with this model indicated that the mechanism of chemical transfer into bedding planes and comminution of coal is dominantly a capillary-imbibition-induced flow phenomenon and to a lesser extent a diffusion-controlled process. Civan and Knapp (1988) also tested this hypothesis using the experimental data of Mamaghani et al. (1987). As reported by Mamaghani et al. (1987), the maximum comminution rates for the middle Pennsylvania Cherokee C-bituminous coal with NaOH solutions were within the range of 6–8% caustic concentration. Hence, Mamaghani (1988) concluded that this contradicts Table 2 of our earlier work (1988), which reported that capillary-imbibition number (reciprocal of the surface-tension number) decreases with increasing caustic concentration. This conclusion is misleading because this table (Civan and Knapp, 1988) simply presents a set of data on capillary-imbibition number vs. NaOH concentration, and it alone cannot determine the caustic concentration for the maximum comminution rate. This rate can be determined by the following correlation method.

The spontaneous comminution of coal is perceived to progress by imbibition of chemical through the natural fractures and bedding, and weakness planes of coal followed by the surface diffusion and/or reaction of chemicals with the coal-bonding substances to dissolve and/or break the bonding substances and disintegrate coal sequentially through the natural fractures and bedding, and weakness planes. Since coal can be viewed as a porous medium, the rate of chemical comminution, R, is assumed to be a function of the pertinent physicochemical properties of the chemical, surface tension, σ, density, ρ, viscosity, μ, and effective mass diffusivity, D^e, and the mean pore diameter, d, of the chemical flow path, which depends on the coal type and its microstructure. A dimensional analysis of these quantities reveals that three dimensionless groups can be formed. These are the comminution, surface-tension, and Schmidt numbers given, respectively, by:

$$N_c = Rd/\mu \qquad (1)$$

$$T_s = \mu^2/(\sigma \rho d) \qquad (2)$$

and

$$Sc = \nu/D^e \qquad (3)$$

In Eq. 3, $\nu = \mu/\rho$ is the momentum diffusivity or kinematic viscosity. D^e is the effective mass diffusivity of chemical into the bonding substance within the natural bedding planes and fractures of coal. Hence, the specific pore surface and the type of bonding substance are the important factors. Note that the effects of temperature and pressure are considered since the chemical properties depend on temperature and pressure. If there are sufficient experimental data, an empirical correlation between the three dimensionless numbers given by Eqs. 1–3 can be developed by an appropriate regression method. The following simplified approach will be taken that considers the difficulties of obtaining accurate data during coal comminution. These difficulties include the measurement of comminution rate and particle size, variations in coal samples taken from the same source, and chemical interactions, assumed small, between chemicals and coal constituents.

It is assumed that chemicals can readily react with the small amount of bonding substance to quickly dissolve and break the bond as soon as they reach the bonding substance while proceeding through the capillary channels in coal. The rate of this is controlled by the momentum diffusivity among other factors. Therefore, the first simplification is to omit the mass diffusivity compared to the momentum diffusivity. The justification for this reasoning is also provided by Civan and Knapp (1988) based on their sensitivity studies using a phenomenological model of the coal comminution process. In addition, for a given coal the mean pore diameter is a constant and can be dropped from the analysis. As a result, a simplified

Table 1. Comminution of Croweburg Bituminous Coal by Various Chemicals

Chemical	Comminution Rate, R, g/cm^2·h (Civan et al., 1987)	$\sigma\nu^{-1}$, 10^3 g/cm^2·s
NH_3	6.6	9.6
CF_3CO_2H	3.0	2.6
HNO_3	1.6	0.81
CH_3SO_3H	0.44	0.71
H_2SO_4	0.55	0.41

correlation method can be developed by carrying out a dimensional analysis between the comminution rate, R, and the surface tension, σ, density, ρ, and viscosity, μ, of the chemical. Only one dimensionless group is obtained, given by:

$$\Pi = R\nu/\sigma \tag{4}$$

Therefore, the experimental data can be correlated to obtain a function of this group as:

$$f_1(R\nu/\sigma) = 0 \tag{5}$$

For convenience, Eq. 5 can be reformulated to express R in terms of $(\sigma\nu^{-1})$ as:

$$R = f_2(\sigma\nu^{-1}) \tag{6}$$

This method is illustrated using the data reported by Civan et al. (1987) for Croweburg bituminous coal of Oklahoma and by Mamaghani et al. (1987) for the high-volatile C-bituminous-type middle Pennsylvania Cherokee group coals. The comminution rates, R, reported by Civan et al. and Mamaghani et al., and the values of $(\sigma\nu^{-1})$ calculated are presented in Tables 1 and 2. These data are plotted on rectangular coordinate paper in Figure 1, which shows that there is a minimum value of $(\sigma\nu^{-1})$, below which comminution does not take place, and that the comminution rate increases with increasing values of $(\sigma\nu^{-1})$ until a limiting value of the comminution rate is reached. This limiting comminution rate is maintained by increased $(\sigma\nu^{-1})$ values, provided that the $(\sigma\nu^{-1})$ value is less than a critical threshold value at which condition the coal almost explodes by instantaneous fragmentation. This threshold value is a characteristic of coal which is, among other factors, a function of the material strength of coal. Hence, the comminution rate until the threshold point can be correlated by:

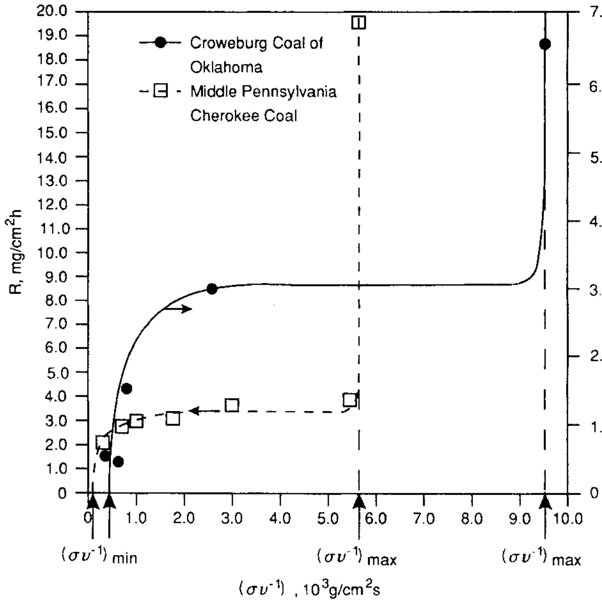

Figure 1. Comminution rate for Croweburg coal of Oklahoma using the Civan et al. (1987) data and middle Pennsylvania Cherokee coal using the Mamaghani et al. (1987) data

$$R = A[1 - \exp\{-B[(\sigma\nu^{-1}) - (\sigma\nu^{-1})_{\min}]\}],$$
$$(\sigma\nu^{-1})_{\min} \le (\sigma\nu^{-1}) < (\sigma\nu^{-1})_{\max} \tag{7}$$

Therefore, a semilog plot of $(1 - R/A)$ vs. $[(\sigma\nu^{-1}) - (\sigma\nu^{-1})_{\min}]$ below the threshold $(\sigma\nu^{-1})_{\max}$ value is linear as shown in Figure 2 for the Civan et al. and Mamaghani et al. data. The parameters of Eq. 7 for Croweburg coal are $A = 3.05$ g/cm^2·h, $B = 1.86 \times 10^{-3}$ (g/cm^2·s)$^{-1}$, $(\sigma\nu^{-1})_{\min} = 5.0 \times 10^2$g/cm^2·s, and $(\sigma\nu^{-1})_{\max} = 9.6 \times 10^3$g/cm^2·s and for the middle Penn-

Table 2. Middle Pennsylvania Cherokee Bituminous Coal by Various Chemicals

Chemical	Comminution Rate, R, mg/cm^2·h (Mamaghani et al., 1987)	$(\sigma\nu^{-1})$, 10^3 g/cm^2·s
Acetone	19.6	5.7
NaOH (7%)	3.9	5.4
Methanol	3.6	3.0
Ethanol	3.1	1.8
n-Propanol	3.0	1.0
n-Butanol	2.8	0.76
n-Hexanol	2.1	0.36

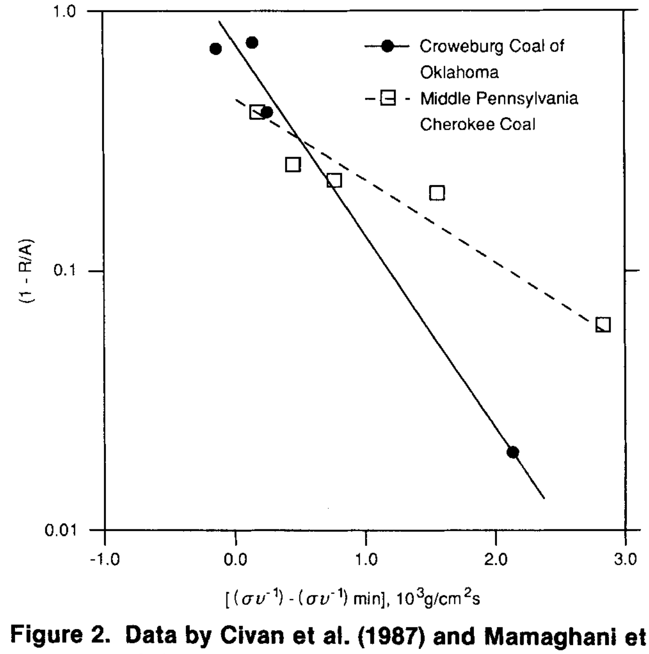

Figure 2. Data by Civan et al. (1987) and Mamaghani et al. (1987).

sylvania Cherokee coal $A = 3.9\ \mathrm{mg/cm^2 \cdot h}$, $B = 7.07 \times 10^{-3}\ (\mathrm{g/cm^2 \cdot s})^{-1}$, $(\sigma\nu^{-1})_{\min} = 20.0\ \mathrm{g/cm^2 \cdot s}$, and $(\sigma\nu^{-1})_{\max} = 5.7 \times 10^3\ \mathrm{g/cm^2 \cdot s}$.

The preceding analysis of the chemical comminution of bituminous coals from two different sources reveals that the comminution processes for both coals suggest a similar mechanism. The inherent assumption that capillary imbibition is the governing factor appears to be valid at least for Croweburg coal of Oklahoma and for the middle Pennsylvania Cherokee coal. Figure 1 revealed that for a given coal there are two critical values. The first is a minimum value of the $(\sigma\nu^{-1})$ value, below which chemical comminution does not occur. The second is a threshold $(\sigma\nu^{-1})$ value beyond which chemical comminution occurs almost instantaneously. This value probably corresponds to the condition at which the internal pressure of coal created by the chemical imbibition exceeds the failure strength of the coal. Between these limits the comminution rate obeys a semilogarithmic law whose parameters depend on the type of coal.

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Notation

A = empirical constant $[ML^{-2}T^{-1}]$
B = empirical constant $[M^{-1}L^2T]$
D^e = effective mass diffusivity $[L^2T^{-1}]$
f_1 = convention for a function
f_2 = convention for a function
d = mean pore diameter $[L]$
T_s = surface-tension number [1]
R = comminution rate $[ML^{-2}T^{-1}]$
N_c = comminution number [1]
Sc = Schmidt number [1]

Basic dimensions

[1] = dimensionless
$[M]$ = mass
$[L]$ = length
$[T]$ = time

Greek letters

μ = viscosity of chemical $[ML^{-1}T^{-1}]$
ν = momentum diffusivity of chemical $[L^2T^{-1}]$
Π = dimensionless number given by Eq. 4 [1]
ρ = density of chemical $[ML^{-3}]$
σ = surface tension of chemical $[MT^{-2}]$

Literature Cited

Keller, Jr., D. V., and C. D. Smith, "Spontaneous Fracture of Coal," *Fuel*, **55**, 273 (1976).

Civan, F., R. M. Knapp, A. P. Hagen, and M-K Wang, "Solvent Selection for Coal Comminution," *Alternative Energy Sources VII*, Hydrocarbons/Energy Transfer, T. Veziroglu, ed., Hemisphere, **5**, 71 (1987).

Civan, F., and R. M. Knapp, "A Phenomenological Model for Solvent Comminution of Coal," *Alternative Energy Sources VIII*, Research and Development, T. Veziroglu, ed., Hemisphere, **2**, 173 (1989).

Civan, F., and R. M. Knapp, "Letter to the Editor," *AIChE J.*, **34**(1), 171 (1988).

Mamaghani, A. H., "Reply," *AIChE J.*, **34**(1), 171 (1988).

Mamaghani, A. H., J. K. Beddow, and A. F. Vetter, "Chemical Comminution of Coal," *AIChE J.*, **33**(2), 319 (1987).

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