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The development of rate expressions and the evaluation of reactivity for gasification of various coal chars with steam and oxygen

A fundamental investigation into the gasification of chars, which were prepared under similar conditions from 19 different coals of fuel ratio ranging from 1.2 [brown coal] to 7.9 [anthracite], was made thermogravimetrically at 850°C using steam ($H_2O(24\%) + N_2$) and at 900° C using oxygen (O_2 (10%) + N_2). The fractional gasification f, based on the assumption that the rate constant k varied as the gasification progressed, was related to the time of gasification θ, according to the expression $f = 1 - exp(-a\theta^b)$, thus being independent of the coalification rank, the gasification conditions, and the sigmoidal character of the $f - \theta$ curves. The average rate constant $\overline{k} = \int_0^1 k\,(f)df$, which was approximately equal to the value of k at $f = 0.5$), and the time of gasification θ necessary to reach the point at which $f = 0.5$ proved to be useful parameters for evalutating the gasification reactivities of chars, and were closely correlated with the fuel ratio of the coal and the atomic ratios H/C of the coal and/or char. The gasification-rate ($f - \theta$) curves of the 19 coal chars were correlated by the equation, $f = 1 - exp(-A\tau^B)$, where $\tau = \theta/\theta_{f=0.5}$.

Introduction

No satisfactory correlation between the rate of gasification of a coal and those of its properties on which this depends has yet been found, although a number of different rate equations have been put forward [1-3, 13]. These, however, have all been empirical equations, require tedious analytical procedures and lack basic physical or chemical significance. In this article, the authors report on a fundamental experimental investigation of gasification with steam and oxygen at constant temperature, of the chars formed from 19 Japanese and foreign coals with different fuel ratios, ranging from brown coal to bituminous coal to anthracite. Curves of fractional gasification *versus* time are analyzed, and a procedure for evaluating reactivity for gasification is proposed, together with a precise yet simple rate equation, which is independent of

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the type of coal and the sigmoidal character of the gasification curves, that is the presence of a maximum in the rate of gasification. It is also shown that the gasification reactivity correlates well with the fuel ratio and also with the H/C atomic ratio of the coal and/or char.

1. Coal samples

Table 1 lists the coals used and their analyses. The ultimate analyses are dry ash-free (daf), C and H being determined by Pregl's method and N by Kjeldahl's method; the remainder was assumed to be O, and S was not determined. The volatile matter (VM) was calculated from the loss in weight when the coal was heated in a nitrogen stream (300 Ncm^3/min) in a thermal balance from 110° C to 900 ° C at a constant rate of 20° C/min, and then held at 900° C for 30 min. The fixed carbon (FC) and the high-temperature ash (HTA) were calculated from the loss in weight, when the residue from the determination of the volatile matter was held at 900° C in a stream of O_2 (10%)-N_2 (300 Ncm^3/min). The amount of volatile matter was almost the same as obtained by heating the samples for 7 min at 900° C in a nitrogen stream (300 Ncm^3/min) in a horizontal tubular furnace (JISM 8812), as well as that obtained by further heating the samples for one hour at 900° C. The low-temperature ash (LTA) was the mineral content of the coal, as determined on a Yanagimoto Seisakusho LTA 2SN oxygen-plasma low-temperature ash apparatus. The caking characteristics of the coals were evaluated visually by observing the states of fusion and expansion of the samples after one hour's heating at 800° C in a stream of nitrogen (300 Ncm^3/min).

The mean particle size of the coal samples was 0.5 mm. Sometimes samples with particle sizes of < 0.5 mm (48 mesh), 1.0 mm, 2.0 mm, and 2.8 mm were used, but there was no change in the HTA or LTA analyses. The dry weights of the coal samples were measured after one hour's treatment at 110° C in a nitrogen stream (dry basis, db). In the graphs which appear in this article, the coal samples will be identified by the letters in the first column of Table 1.

2. Experiments

The apparatus and procedure in the gasification experiments were the same as in our earlier reports [7,9]. Simultaneous measurements were made of the exit gas composition (H_2, CO, and CO_2) by gas chromatography, and of the gasification rate using a conventional flow-type thermal balance. In the

Table 1. Coal samples employed.

Coal		Country of Origin	Caking	Ultimate analysis [wt%, daf]				Atomic ratio			Proximate analysis [wt%, db]				Fuel ratio
				C	H	N	O_{diff}*	$\frac{H}{C}$	$\frac{O_{diff}*}{C}$	$\frac{N}{C}$	Volatile matter: VM	Fixed carbon FC	Ash (HTA)	$\frac{LTA}{HTA}$ [—]	$\left(FR=\frac{FC}{VM}\right)$ [—]
H	Horonai	Japan	moderate	79.34	6.89	1.63	12.14	1.03	0.116	0.018	31.61	38.57	29.82	1.05	1.22
M	Miike	Japan	strong	83.91	6.51	1.22	8.36	0.92	0.075	0.013	39.01	53.71	7.28	1.41	1.38
Co	Collie	Australia	none	90.46	4.64	1.37	23.53	0.79	0.251	0.017	36.37	59.57	4.06	2.09	1.64
A	Abersee	Australia	moderate	79.54	5.71	2.02	12.73	0.86	0.121	0.022	33.12	56.81	10.06	1.13	1.72
L	Lithgow	Australia	moderate	82.14	5.66	1.88	10.32	0.82	0.094	0.020	31.74	57.73	10.53	1.32	1.82
W	Woodland	Australia	moderate	82.75	5.84	1.72	9.69	0.84	0.089	0.018	31.32	57.88	10.80	1.04	1.85
KM	Kellerman	U.S.A.	strong	82.61	5.88	1.79	9.72	0.86	0.088	0.019	34.51	64.47	1.02	1.65	1.87
Ma	Matla	R.S. Africa	none	77.72	5.91	1.93	14.44	0.91	0.139	0.021	31.16	61.72	7.12	1.32	1.98
Z	Zontagsvlei	R.S. Africa	weak	83.89	5.80	2.05	8.26	0.83	0.074	0.021	30.23	63.36	6.41	1.45	2.10
BA	Blair Athol	Australia	none	78.58	5.32	1.64	14.46	0.81	0.139	0.017	27.20	62.65	10.15	1.07	2.30
BW	Black Water	Australia	strong	79.35	5.12	1.95	13.58	0.77	0.129	0.021	26.97	65.62	7.41	1.49	2.43
Ca	Callide	Australia	none	75.55	4.80	1.12	18.53	0.76	0.184	0.013	26.65	65.91	7.44	1.85	2.47
T	Tatung	P.R. China	moderate	85.41	5.19	0.90	8.50	0.72	0.075	0.009	25.89	64.79	9.32	1.18	2.50
B	Balmer	Canada	strong	83.68	4.62	1.16	10.54	0.66	0.028	0.012	22.08	69.08	8.84	1.29	3.13
P	Pittstone	U.S.A.	strong	85.62	4.07	1.34	8.97	0.57	0.078	0.013	19.32	77.41	3.27	1.32	4.00
S	Smoky River	Canada	strong	86.36	4.47	1.17	8.00	0.62	0.028	0.012	17.84	75.11	7.05	1.67	4.21
Ky	Keystone	U.S.A.	moderate	91.16	4.81	1.32	2.71	0.63	0.022	0.012	15.42	79.00	5.58	1.62	5.12
Ku	Kuznetsk	U.S.S.R.	moderate	88.06	4.28	1.72	5.94	0.58	0.051	0.017	12.95	78.99	8.07	1.18	6.10
U	Uonuki	Japan	none	89.56	4.49	1.69	4.26	0.60	0.036	0.016	10.13	80.05	9.82	1.21	7.91

* $O_{diff} = 100-(C+H+N)$

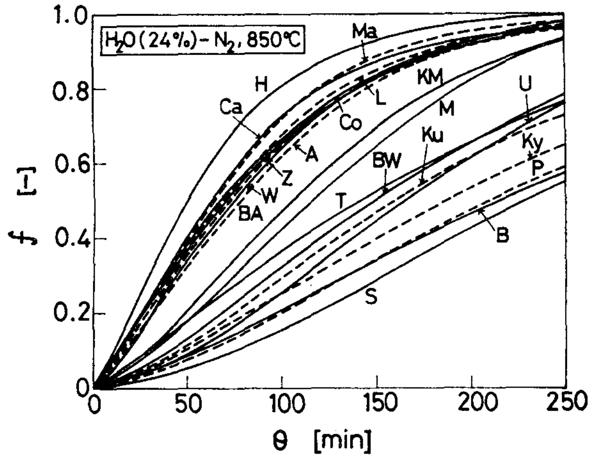

Fig. 1. Rate of gasification of coal char with steam.

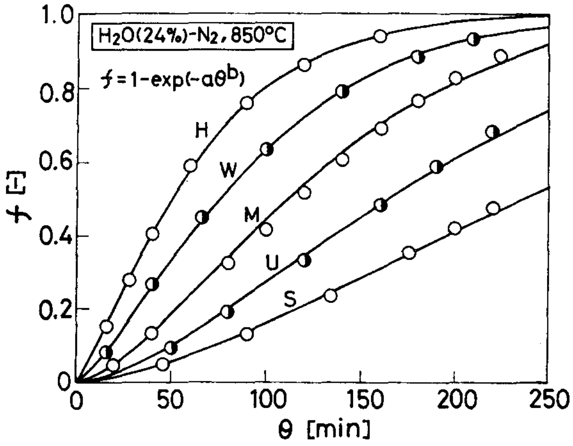

Fig. 3. Fitting of data shown in Fig. 1. to Eq. (1).
Solid lines: Calculated from Eq. (1) using the parameters, *a* and *b*, determined by least squares.

steam-gasification experiments, a reaction tube of 16.5 mm internal diameter and a 13.5-mm-diameter platinum sieve basket were used, with automatic recording, while in the oxygen gasification experiments, a reaction tube of 29 mm internal diameter and a 15-mm-diameter platinum perforated basket were used, with manual operation.

The steam-gasification experiments were carried out at 850° C in a gas stream (H_2O (24%)-N_2, overall flow rate 400 Ncm^3/min), using the char (100 mg, mean particle size 1.0 mm) which was produced by heating the coal for one hour at 900° C in a stream of nitrogen to drive off the volatile matter. The oxygen-gasification experiments were carried out at 900° C in a gas stream (O_2 (10%)-N_2, overall flow rate 300 Ncm^3/min), using the char remaining after measuring the volatile matter by heating the coal (200 mg, mean particle size 0.5 mm). Both types of gasification experiments were also carried out using some of the coal samples, with the particle size being varied over the range 0.5-2.8 mm, but the gasification rates were found to be virtually unaffected by any such change in the particle size.

3. Experimental results and discussion

3.1. Gasification rate equation and method of evaluating the reactivity of the char

3.1.1. Gasification rate equation

The fractional gasification *f* was defined as the fractional weight loss due to the gasification of the fixed-carbon content of the proximate analysis,

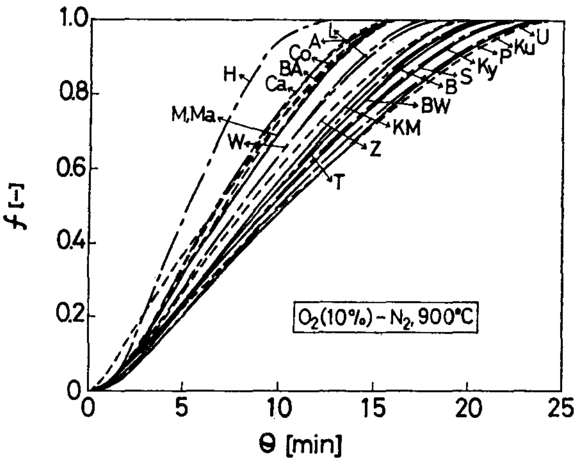

Fig. 2. Rate of gasification of coal char with oxygen.

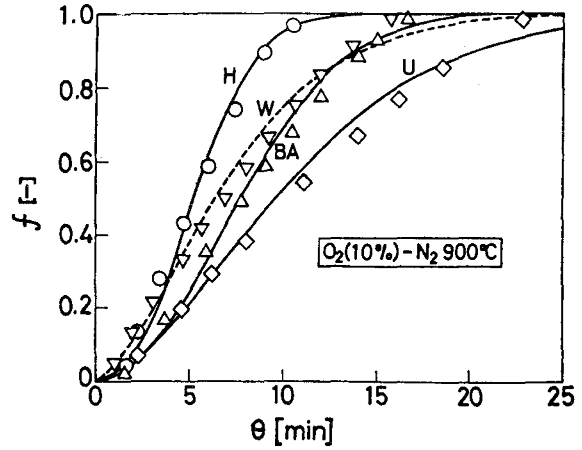

Fig. 4. Fitting of data shown in Fig. 2. to Eq. (1).
Solid lines: Calculated from Eq. (1) using the parameters, *a* and *b*, determined by least squares.

that is, the dry, ash-free content of the char. Figures 1 and 2 show the relationships between the fractional gasification f and the gasification time θ for steam and oxygen gasification, respectively, of the 19 coal chars. It can be seen that, when similarly prepared chars were gasified under similar conditions, the gasification proceeded differently depending on the type of coal used, with some chars exhibiting a distinct sigmoidal character, that is, the existence of a maximum gasification rate $df/d\theta$ somewhere in the range $0 < f < 1$, and others exhibiting no such sigmoidal character.

It is widely recognized [1] that the same coal char can exhibit sigmoidal character or not, depending on the gasification temperature and the composition of the gaseous atmosphere [7,11] and whether or not a catalyst is being used [7,9]. However, the models for the reaction between a solid and a gas which have been put forward to-date, such as the continuous model [5] and the unreacted-core model [4], are incapable of predicting those gasification rates for which a sigmoidal character will be present. Also, the essential nature of sigmoidal character is not clear. In a qualitative sense, the changes in the chemical structure resulting from oxidation and decomposition of the carbonaceous substances and hydrocarbons (aromatic substances, *etc.*) and from dehydration, decomposition, and oxidation of the minerals (hydrates, carbonates, sulfides, *etc.*) [12,14] and the increases in the surface area due to pore formation and the decreases therein due to pore expansion [8,11], together with the changes in the physical structure resulting from increases in the exposed proportions of the surface areas of the catalytically active minerals and ash, as the internal surface areas expand due to the effects just mentioned, result in a fairly complicated overall effect.

Figures 3 and 4 show the result of fitting the very simple two-constant integral rate equation which we introduced in [9],

$$f = 1 - \exp(-a\theta^{b}); \quad \text{where } a \text{ and } b \text{ are constants} \tag{1}$$

to some of the $f - \theta$ data from Figures 1 and 2, respectively. An excellent fit was obtained.

The constants a and b are readily calculated by the least-squares method from the linearized form of Equation (1) as $Y = \ln a + bX$, where $Y = \ln[-\ln(1 - f)]$ and $X = \ln\theta$. The experimental points in Figures 3 and 4 are typical of those which were used in the determination of a and b.

A simple analysis of Equation (1) reveals that, when $0 < b \leqq 1$, there is no sigmoidal character, the rate of gasification decreasing with time. When $b = 1$, Equation (1) is identical with the equation for chemical-reaction rate control in the continuous model [5], with a then being the rate constant. If $b > 1$, a sigmoidal character occurs, and the values of θ and f corresponding to the inflection point or point of maximum gasification rate on the $f - \theta$ curve are given by the equations,

$$\theta_{\text{Inf.}} = [(b-1)/ab]^{1/b} \tag{2}$$

$$f_{\text{Inf.}} = 1 - \exp[-(b-1)/b] \tag{3}$$

The values of $f_{\text{Inf.}}$, as calculated from Equations (2) and (3), were 0.14-0.31 for steam gasification (Figures 1 and 3), and 0.28-0.44 for oxygen gasification (Figures 2 and 4).

3.1.2. Method of evaluating the reactivity

In [6], the authors have used the unreacted-core model to examine the case in which the rate constant and the effective diffusion coefficient in the solid product vary with the position within the solid particles (that is, when they are distributed nonuniformly), and have found on theoretical grounds that the $f - \theta$ curve has a sigmoidal character when the reactivity or rate constant is nonuniformly distributed (as a general rule, the rate constant increases toward the center of the solid particles). If the rate constant is constant throughout the particle, there is no sigmoidal character, even if the effective diffusion coefficient, which is closely dependent on the physical structure, is distributed nonuniformly.

On the basis of the assumption that the rate constant does not remain constant throughout the reaction, Gardner *et al.* [2], Johnson [3], and Rai *et al.* [13] made attempts at a rate analysis, by assuming that the gasification rate $df/d\theta$ of coal char is proportional to the fraction of unreacted char $(1 - f)$ to the power 1 to 2/3, and that the energy of activation is a function of f, thus:

$$E = E_0 + E_{01}f + E_{02}f^2; \quad E_0, E_{01}, E_{02}: \tag{4}$$

where E_0, E_{01}, and E_{02} are constants. However, they were unable to obtain analytically an integral rate equation. The required analysis of the data is rather tedious, and there is no rationale for their treatment.

If, as an index of reactivity, the rate constant itself is assumed to vary with the gasification time

θ, and thus with f, and the gasification rate is assumed to be directly proportional to the fraction of unreacted char $(1-f)$, then

$$df/d\theta = k(\theta)(1-f) \tag{5}$$

which, if integrated, gives

$$f = 1-\exp\left[-\int_0^{\theta} k(\theta)d\theta\right] \tag{6}$$

Comparing Equations (1) and (6), we can easily see that the variation in the rate constant corresponding to Equation (4) can be modeled by the expressions,

$$k(\theta) = ab\theta^{b-1} \tag{7}$$

and

$$k(f) = a^{1/b}b[-\ln(1-f)]^{(b-1)/b} \tag{8}$$

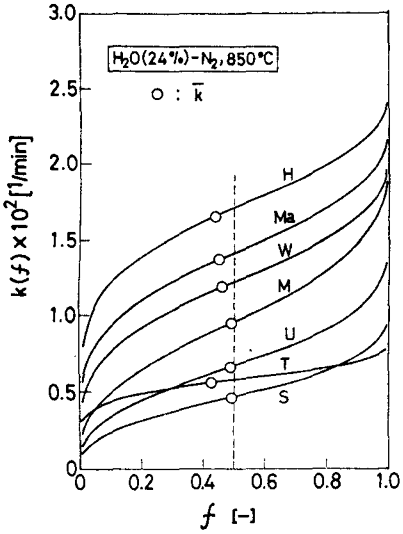

Fig. 5. Variation of rate constant ($k(f)$) calculated from Eq. (8) for gasification of coal char with steam.

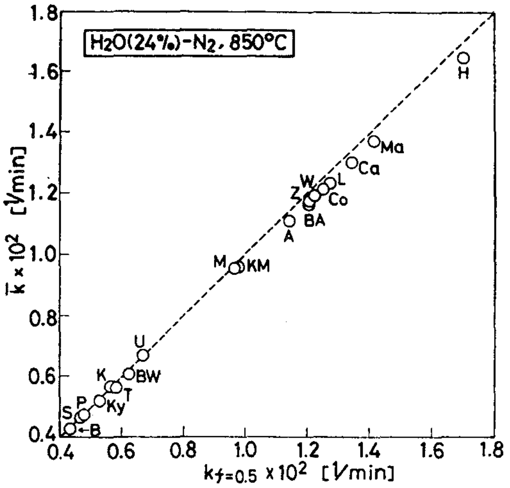

Fig. 6. Approximation of average rate constant ($\bar{k}$) with rate constant at $f=0.5$ ($k_{f=0.5}$) for gasification of coal char with steam.

Figure 5 shows some typical curves for the variation in the rate constant k over the entire gasification process, as calculated from Equation (8), using the values of a and b obtained by data fitting in the rate equation in Figure 3. Thus, when sigmoidal character was in evidence, k increased as the gasification progressed, while in the case of the Tatung coal which exhibited no clear sigmoidal character (see Figure 1), k showed an almost constant distribution. Cases are also known where the addition of a catalyst causes the rate constant to decrease as the gasification progresses [9]. The open circles which have been plotted in Figure 5 represent the average rate constant $\bar{k}$, as calculated from the following expression (by numerical integration from $f = 0.01 - 0.99$ using Simpson's formula):

$$\bar{k} = \int_0^1 k(f)df \tag{9}$$

The values of $\bar{k}$ are fairly close to the rate constants corresponding to $f = 0.5$, $k_{f=0.5}$, thus:

$$k_{f=0.5} = a^{1/b}b[\ln 2]^{(b-1)/b} \doteqdot \bar{k} \tag{10}$$

Figure 6 shows the fit obtained by applying Equation (10) to the data for steam gasification of the 19 coal chars under study. After including the results for oxygen gasification, the error of the fit was within about ± 3%, which means that $\bar{k}$, or

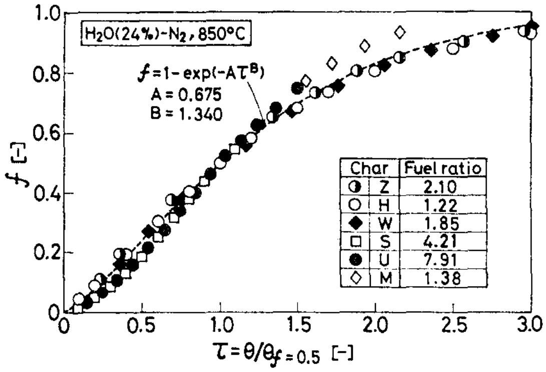

Fig. 7. Fitting of data for gasification of each coal char with steam to Eq. (11).

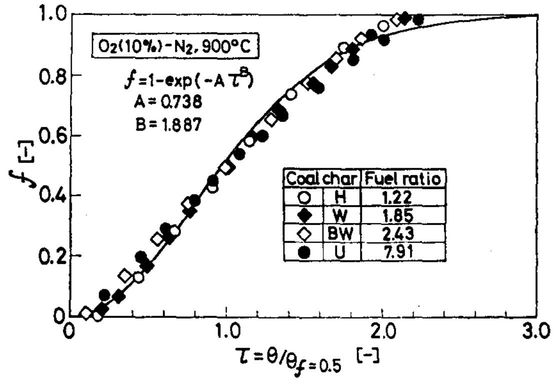

Fig. 8. Fitting of data for gasification of each coal char with oxygen to Eq. (11).

$k_{f=0.5}$, is a suitable criterion with which to evaluate and compare quantitatively the effects on gasification reactivity of the addition of a catalyst [9] or of the gasification conditions [9].

3.2. Correlation of the gasification rate

Figures 7 and 8 show, for some of the 19 coal chars studied, the relationship between the fractional gasification f and the dimensionless gasification time $\tau(=\theta/\theta_{f=0.5})$, standardized with respect to the gasification time $\theta_{f=0.5}$ at $f=0.5$ [11]. It can be seen that, both for steam gasification (Figure 7, based on Figure 1) and for oxygen gasification (Figure 8, based on Figure 2), the relationship was virtually independent of the coal type, the results being represented by a single sigmoidal curve. This is a similar result to that of Mahajan *et al.* [11], who performed a similar type of experiment*.

The sigmoidal curves in Figures 7 and 8 were plotted using the equation,

$$f=1-\exp(-A\tau^{B});\quad \tau=\theta/\theta_{f=0.5} \qquad (11)$$

which is identical with Equation (1), with the constants A and B being determined from the data for all 19 coal chars. The correlating coefficients were 0.995 in the case of steam gasification and 0.980 in the case of oxygen gasification.

For gasification with either steam and oxygen, there was a good correlation (shown in Figure 9) between $\theta_{f=0.5}$ and the average rate constant $\bar{k}$ as calculated from Equation (9) and the rate Equation (1), thus providing us with another criterion for evaluating gasification reactivities, namely $\theta_{f=0.5}$. Equation (11) can therefore be used for design purposes, since, if we determine $\theta_{f=0.5}$ experimentally, it is possible to use Equation (11) (the equation for the curves in Figures 7 and 8) to estimate the value of θ for any value of f.

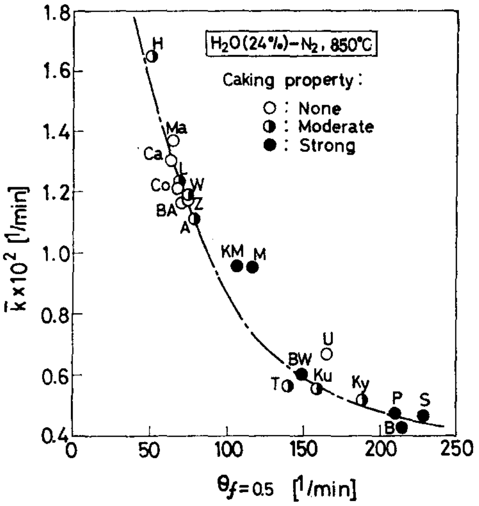

Fig. 9. Relation between average rate constant ($\bar{k}$) and time of gasification $f=0.5$ ($\theta_{f=0.5}$) in gasification of each coal char with steam.

The observation that the shape of the gasification curve is independent of the coal type suggests that the mechanism of gasification is basically the same for all types of char.

On a dry basis, the gaseous product from the gasification with oxygen under the above conditions was chiefly CO_2, with the concentration of CO being less than about 2%; in gasification with

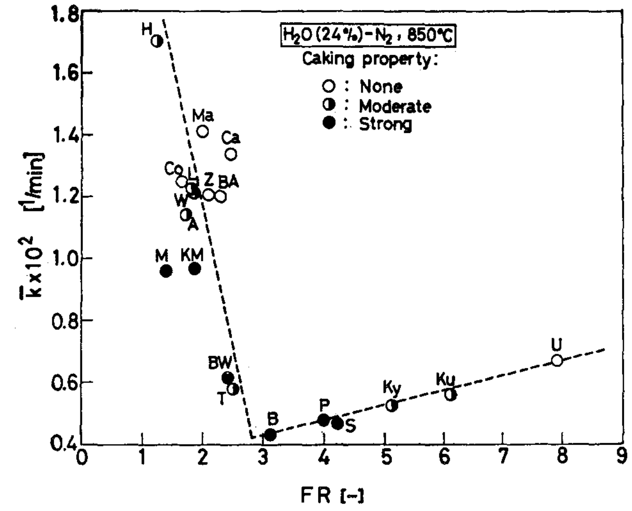

Fig. 10. Relation between average rate constant ($\bar{k}$) and fuel ratio (FR) in gasification of each coal char with steam.

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*Mahajan *et al.* [11] attempted to correlate the rates of gasification by air, steam, carbon dioxide, and hydrogen, using the three-constant rate expression, $f=A\tau+B\tau^2+C\tau^3$, but with such an empirical equation, the constants did not have any physical significance.

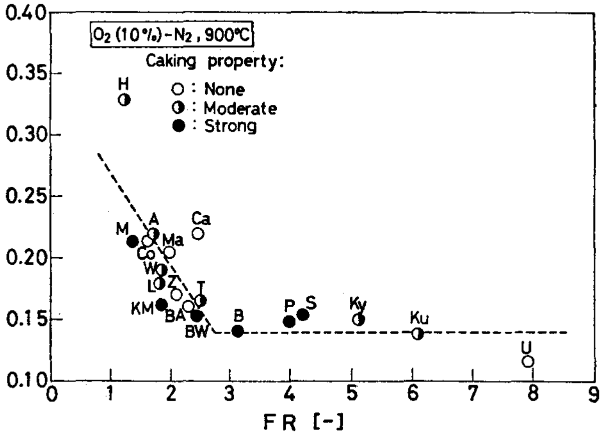

Fig. 11. Relation between average rate constant ($\bar{k}$) and fuel ratio (FR) in gasification of each coal char with oxygen.

steam, the gaseous product was composed of H_2, CO, and CO_2, the mole ratio $H_2/(CO + CO_2)$ being 1.1-1.6. When we contrast this with our previous experimental findings [7,9], namely that, when pure ash-free carbon, prepared by pyrolysis of a synthetic organic resin, the copolymer poly(vinylidene chloride) 70%-poly(vinyl chloride) 30%, in a stream of nitrogen, was gasified with steam, with the temperature being raised from ambient to 900° C at the rate of 4° C/min, there was a binary gaseous reaction product with a H_2/CO mole ratio of ~ 1, we are led to the conclusion that, as the main reaction product from any type of coal char is H_2, the mineral and ash contents of the coal catalyze the conversion of CO according to the reaction, $CO + H_2O \rightarrow CO_2 + H_2$.

3.3. Relationship between the properties of the coal and the gasification reactivity of the char

Figures 10 and 11 show, for the gasification-rate data for steam and oxygen, respectively, the correlation between the reactivity $\bar{k}$, as calculated using the analytical procedure described above, and the fuel ratio, as calculated from the proximate analysis given in Table 1. In both cases, $\bar{k}$ correlated well with the fuel ratio FR of the coal. The gasification reactivity behaved quite differently on either side of a fuel ratio of ~ 3. When $FR \gtrsim 3$, the gasification reactivity was only slightly dependent on the fuel ratio (or coalification rank) in the case of gasification with steam or was virtually independent of it in the case of gasification with oxygen. When $FR \lesssim 3$, the gasification reactivity varied rapidly and almost linearly with the fuel ratio.

Figure 12 shows the relationship between $\bar{k}$ and the atomic ratio H/C of the coal. In spite of the considerable scatter, there is a reasonable correlation, the correlation coefficients being 0.85 for gasification with steam and 0.80 for gasification with oxygen.

It has been shown elsewhere that there is no correlation between $\bar{k}$ and the carbon content of the coal (C) [10], which has been used in the past as a criterion representing the properties of a coal, the correlation coefficients being 0.67 for gasification with steam and 0.57 for gasification with oxygen. Although the relevant graphs have not been illustrated, the correlations between the atomic ratio H/C of the coal char and $\bar{k}$ were similar to those for the coal as shown in Figure 12, from which we can deduce that, even after the coal had been charred, its basic structure was preserved.

Conclusions

Fundamental experiments were carried out on gasification with steam (H_2O (24%)-N_2) and oxygen (O_2 (10%)-N_2) of chars, which were prepared from 19 coal samples ranging from brown coal to anthracite (fuel ratio FR = 1.2-7.9). The gasification rate equation was determined and methods of evaluating the reactivity were examined, and the following results were obtained:

1) Rate Equation (1), which assumes that the

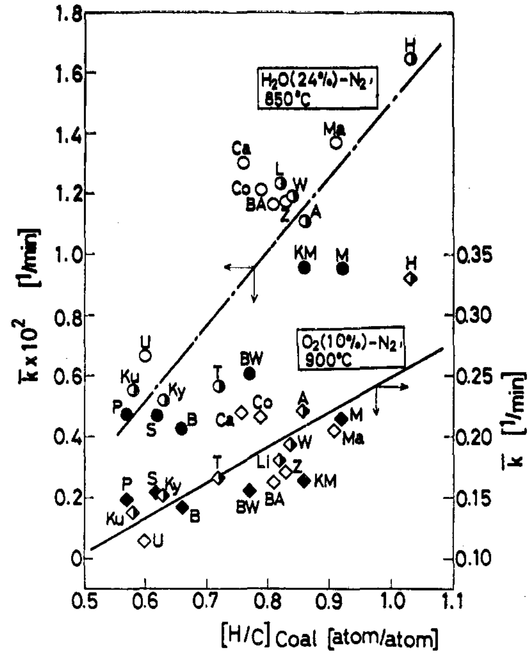

Fig. 12. Correlation between average rate constant ($\bar{k}$) and H/C atomic ratio of coal for gasification of each coal char with steam and oxygen.

rate constant k varies with θ or f according to Equation (7) and (8) is a fairly precise expression for the relationship between the fractional gasification f and the gasification time θ. Equation (1) could be adapted to the data for f *versus* θ, independent of the type of coal, the gasification conditions, and whether or not a sigmoidal character was present.

2) It was possible to evaluate and compare quantitatively the gasification reactivities $\bar{k}$ of coal chars on the basis of the average rate constant $\bar{k}$ (Equation (9)) or the rate constant at $f = 0.5$, $k_{f=0.5}$. The gasification time required to reach the point where $f = 0.5$, $\theta_{f=0.5}$, can also be used as a criterion for evaluating the reactivity.

3) When the dimensionless gasification time τ ($= \theta/\theta_{f=0.5}$), standardized on $\theta_{f=0.5}$, was used in place of the gasification time θ, the relationship between f and τ for the 19 coal chars correlated precisely using Equation (11), being a single sigmoidal curve characteristic of the conditions of gasification.

4) When the fuel ratio FR was between 1 and 3, the different coals exhibited widely different reactivities $\bar{k}$ for gasification. Low-coalification-rank coals, for which FR was low, gave chars with high reactivities for gasification, whereas with chars produced from high-coalification-rank coals for which FR $\gtrsim 3$, the reactivity for gasification was independent of the coal type.

5) The relationship between the atomic ratio H/C of a coal or char and the fuel ratio of coal FR corresponded to the relationship between $\bar{k}$ and *FR*; the reactivity $\bar{k}$ to gasification correlated well with the atomic ratio H/C of the coal or char.

Acknowledgement

This research was supported in part by a grant in aid for scientific research from the Ministry of Education.

Nomenclature

A, B	parameters in characteristic rate Equation (11), [−]
a, b	parameters in gasification rate Equation (1), [−], $\min^{-b}$
(C)	carbon content of coal, weight % (dry, ash-free)
E, E_0	energy of activation defined by Equation (4), J/K·mol
E_{01}, E_{02}	parameters in Equation (4), J/K·mol
FC	fixed-carbon content of coal, weight % (dry basis)
FR	fuel ratio (= FC/VM)
f	fractional gasification of coal char, (g FC gasified)/(g FC)
$f_{\mathrm{Inf.}}$	value of f at point of inflection or point of maximum rate of gasification on $f - \theta$ curve
[H/C]	hydrogen/carbon atomic ratio
$k(f)$	rate constant dependent on f, defined by Equation (8), $\min^{-1}$
$k(\theta)$	rate constant dependent on θ, defined by Equations (5) and (7), $\min^{-1}$
$\bar{k}$	average rate constant defined by Equation (9), $\min^{-1}$
$k_{f=0.5}$	rate constant at $f = 0.5$, $\min^{-1}$
VM	volatile-matter content of coal, weight % (dry basis)
θ	time of gasification, min
$\theta_{\mathrm{Inf.}}$	value of θ at point of inflection or point of maximum rate of gasification on $f - \theta$ curve, min
$\theta_{f=0.5}$	gasification time when $f = 0.5$, min
τ	dimensionless time of gasification ($= \theta/\theta_{f=0.5}$)

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