RELATIONSHIP BETWEEN COAL CHARACTERISTICS AND ITS REACTIVITY ON HYDROLIQUEFACTION

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INTRODUCTION

The reactivity of coal during hydroliquefaction should be related to the characteristics of coal as are determined by ultimate, proximate and petrographic analyses, etc.

The relationship between the characteristics of coal and its reactivity has been studied by several researchers. As a result, several typical parameters are purported to be in this relationship (1,2,3). Among these parameters, carbon content, mean reflectance, volatile matter, H/C atomic ratio, reactive macerals, etc. are reported to be related to coal reactivity. However, it is generally known that these are usually applied only in limited conditions of hydroliquefaction. Therefore, attempts to find better parameters still continue.

In this study the most closely related parameter to coal reactivity, as represented by conversion, has been selected by liquefying several types of coals covering a wide range from lignite to bituminous coals as well as samples from a narrow range collected at different mining sites. Selected coals from the wide range of rank are located in the coal band shown in Fig. 1. The resulting parameters are compared with other parameters reported by other papers (2,3).

EXPERIMENTS AND RESULTS

The liquefaction of coals was studied in a 500 ml magnetically stirred stainless steel autoclave. Analytical data on coals used in this study are presented in Tables 1 and 2. Hydroliquefaction data on coals used in this study are summarized in Tables 3 and 4.

The experimental methods and procedures were carried out by conventional methods.

Conversion was calculated as follows.

$$\text{Conversion \%} = \frac{\text{Coal charged (d.a.f.)} - \text{Insoluble residue (d.a.f.)}}{\text{Coal charged (d.a.f.)}} \times 100$$

The relations between coal reactivity and several parameters representing coal characteristics are shown in Figs. 2 to 7. In these figures the reactivity of coal is measured by conversion. In the results, volatile carbon % is selected as a more closely related parameter than the common parameters, such as C %, H %, O %, H/C atomic ratio, volatile matter, etc.

Volatile carbon % is defined by the equation as follows.

$$\text{Volatile carbon \%} = \text{C \% (d.a.f.)} - \frac{\text{Fixed carbon \%}}{\text{Volatile matter \%} + \text{Fixed carbon \%}} \times 100$$

This parameter is derived from the following basis. It is generally considered that the first step of coal hydroliquefaction is the thermal decomposition of C-C and C-O bonds, etc. in coal structure. Thus, it is presumed that the volatile matter in coal is closely related, as a parameter to coal reactivity (conversion). But, the amounts of oxygen containing compounds, such as carbon dioxide, water, etc. in volatile matter vary greatly with the rank of coal. Therefore, the carbon % in volatile matter (Volatile carbon %) will be considered as the parameter representing reactivity.

DISCUSSION

It is well known that coal reactivity depends on the solvent, the conditions of hydroliquefaction, and the composition of the coal. Thus, it is desirable that the parameter representing coal reactivity shows essentially the same tendency, despite the conditions of hydroliquefaction. Accordingly comparison of parameters were carried out, using some previously reported results (2,3).

The following results were obtained as shown in the example below. The relationship between conversion and C % in coal (d.a.f.) is shown in Fig. 2. In this figure, the relatively close relationship between conversion and C % is observed, but at the same time it is found also that there are exceptions in this relationship. The behaviour of abnormal coals could possibly be explained by inert content of the coal at the same carbon level. That is, in Yamakawa's data, the inert content of Miike and Yubari coals are lower, while Griffin coal is higher. The same result can be observed in our results. The inert content of Lithgow coal is high. Furthermore, in P. H. Given's data, the lignite sample, PSOC 87 coal deviated considerably from the general tendency. This seems to indicate that this coal was chemically treated (3). It is found also that the consequences of this relationship depend on the conditions of liquefaction and coal quality used (Fig. 2). Thus, C % in coal is not appreciably useful as a parameter. Similar consequences are found in the relationship between conversion and other parameters, such as H %, O % in coal.

The relationship between conversion and the H/C atomic ratio of coal is shown in Fig. 3. In this figure, a good relationship is found to hold in some restricted conditions of liquefaction, but the consequences of this relationship are not general.

The relationship between conversion and volatile matter % (d.a.f.) in coal is shown in Fig. 4. The consequences of this relationship are found to differ greatly from one another in the conditions of liquefaction and the coals used.

The relationship between conversion and volatile carbon % in coal is shown in Fig. 5. According to this relationship as shown in Fig. 5, conversion of almost all coals in our research can be expressed exclusively under the same experimental conditions. It was further found that the effect of a catalyst was larger in coals of lower volatile carbon %. In Yamakawa's data, a fairly good relationship is found except for abnormal coals of high sulphur content(Kentucky No. 11) and of high inert content (Griffin), though the behaviour of Taiheiyo coal can not be explained. In addition, in Given's data, a similar good relationship is found except for an abnormal coal (PSOC 99).

In spite of the differences of liquefaction conditions and coals used, the consequences of this relationship are almost the same except for some abnormal coals. Therefore, it is safe to say that coals of higher volatile carbon % are more reactive than those of lower volatile carbon %. Thus, volatile carbon % seems to be a better parameter to estimate coal reactivity.

The relationship between conversion and the mean maximum reflectance of coal is shown in Fig.6. In Yamakawa's data, a fairly good relationship is found except for abnormal coals of high inert content (Griffin, Grose valley) and low inert content (Miike). And a similar good relationship is found also in Given's data. However, the consequences of this relationship are the reverse in both cases.

The relationship between conversion and petrographic components % in coal is shown in Fig. 7. In this figure, a good relationship between conversion and reactive macerals % in coals is observed. Furthermore, fairly good relationships between conversion and inert ingredients % in coal are also observed. And the consequences of this relationship are essentially the same in two different liquefaction conditions. Thus, it is concluded that the reactive macerals % or inert ingredients % in coal are better parameters to estimate coal reactivity.

In coals of the narrow range of rank (Morwell brown coal) volatile carbon % and other parameters are in relation to the color tone of brown coal as shown in

Table 2. Thus, the reactivity of Morwell brown coal is roughly represented by the color tone of the coal (lithotype). That is, "Light" coal is more reactive than "Dark" coal.

The experimental results described above seem to demonstrate that volatile carbon % in coal (a new parameter) is very useful as a parameter to estimate coal reactivity. However, further study is necessary to clarify the validity of this parameter.

CONCLUSION

A good correlation between volatile carbon % in coal (a new parameter) and coal reactivity was observed. That is, conversion increases with the increasing volatile carbon %. Further,.the effect of a catalyst is larger in coals of lower volatile carbon %. In addition, a similar good correlation between petrographic components % in coal (reactive macerals %, inert ingredients % in coal) and coal reactivity was confirmed. In coals of the narrow range (Morwell brown coal), a lithotype, distinguishing the color tone of the coal, is valuable as a parameter. "Light" coal is more reactive than "Dark" coal.

ACKNOWLEDGEMENT

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Table 1. Analytical data on coals used in the study of the wide range

Coal quality / Coal type	Ultimate analysis (d.a.f.)							Proximate analysis*				Volatile carbon content%
	C	H	O	N	S	H/C	O/C	Moisture	Ash	Volatile Matter	Fixed Carbon	
Yallourn brown coal	62.4	4.2	32.6	0.6	0.2	0.800	0.392	11.4	0.8	45.5	42.3	14.2
Taiheiyo coal	73.0	5.6	19.4	1.6	0.4	0.912	0.199	6.3	13.6	40.2	39.9	23.2
Lithgow coal	81.5	5.1	11.0	1.6	0.8	0.744	0.101	1.7	12.1	32.6	53.6	19.3
Miike coal	81.5	5.7	8.3	1.3	3.2	0.832	0.076	0.7	13.4	39.8	46.1	27.8

* Equilibrium moisture content

Table 2. Analytical data on Morwell brown coals used in the study of the narrow range

Coal quality / Coal sample	Ultimate analysis (d.a.f.)							Proximate analysis(dry)				Volatile carbon content%	Litho-type
	C	H	O	N	S	H/C	O/C	Moisture*	Ash	Volatile matter	Fixed carbon		
A	67.05	3.70	28.36	0.58	0.31	0.656	0.317	23.3	2.4	49.4	48.2	17.66	Dark
B	69.36	4.12	25.45	0.82	0.26	0.713	0.275	15.2	2.3	45.8	51.9	16.24	Medium Dark
C	69.82	4.45	24.49	0.92	0.32	0.765	0.263	15.7	2.7	47.3	50.0	18.43	Medium Light
D	69.49	4.72	24.79	0.76	0.27	0.815	0.267	14.5	2.9	53.7	43.4	24.79	Light
E	71.85	4.90	21.95	0.87	0.43	0.811	0.229	21.2	2.7	51.2	46.0	24.52	Light

* Equilibrium moisture content

Table 3. Reaction conditions and results of hydroliquefaction on coals used in the study of the wide range

Feed Coal		Yallourn coal		Lithgow coal		Taiheiyo coal		Miike coal	
Feed Coal (g. as d.a.f.)		43.9		43.1		40.1		43.0	
Solvent*		150		150		150		150	
Catalyst (g)	Fe_2O_3	0	0.75	0	0.75	0	0.75	0	0.75
	S	0	0.15	0	0.15	0	0.15	0	0.15
Hydrogen initial pressure (kg/cm^2)		60		60		60		60	
Reaction Temperature (^oC)		450		450		450		450	
Holding Time at Reaction Temp. (hr.)		2		2		2		2	
Conversion** (%)		28.5	55.0	41.8	58.9	60.3	65.2	91.1	97.4

* Solvent consists of creosote oil and recovered solvent.

** Conversion was calculated by benzene insoluble residue.

Table 4. Reaction conditions and results of hydroliquefaction on Morwell brown coal used in the study of the narrow range

Feed coal sample		A	B	C	D	E
Feed coal (g. as d.a.f.)		37.5	37.5	37.5	37.5	37.5
Solvent*		112.5	112.5	112.5	112.5	112.5
Catalyst (g)	Fe_2O_3	0.54	0.54	0.54	0.54	0.54
	S	0.22	0.22	0.22	0.22	0.22
Hydrogen initial pressure (kg/cm^2)		80	80	80	80	80
Reaction Temperature (^oC)		430	430	430	430	430
Holding Time at Reaction Temp. (hr.)		1.0	1.0	1.0	1.0	1.0
Conversion**		59.0	75.8	81.6	87.7	80.8

* Solvent means creosote oil.

** Conversion was calculated by pyridin insoluble residue.

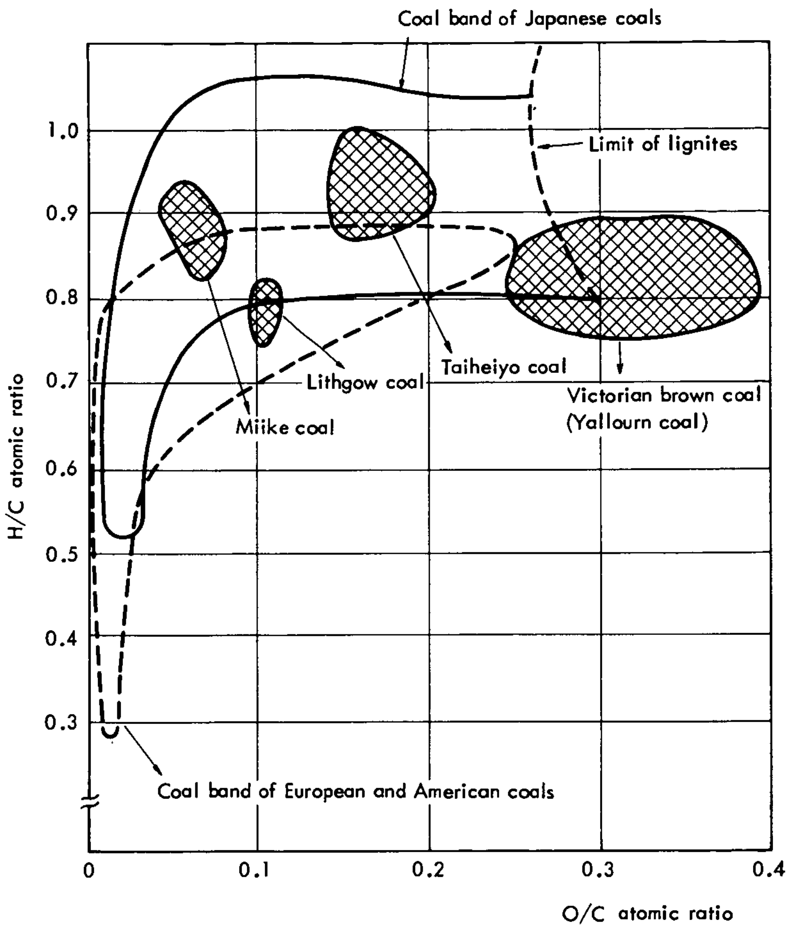

Fig. 1 Relationship between coals used and coal bands

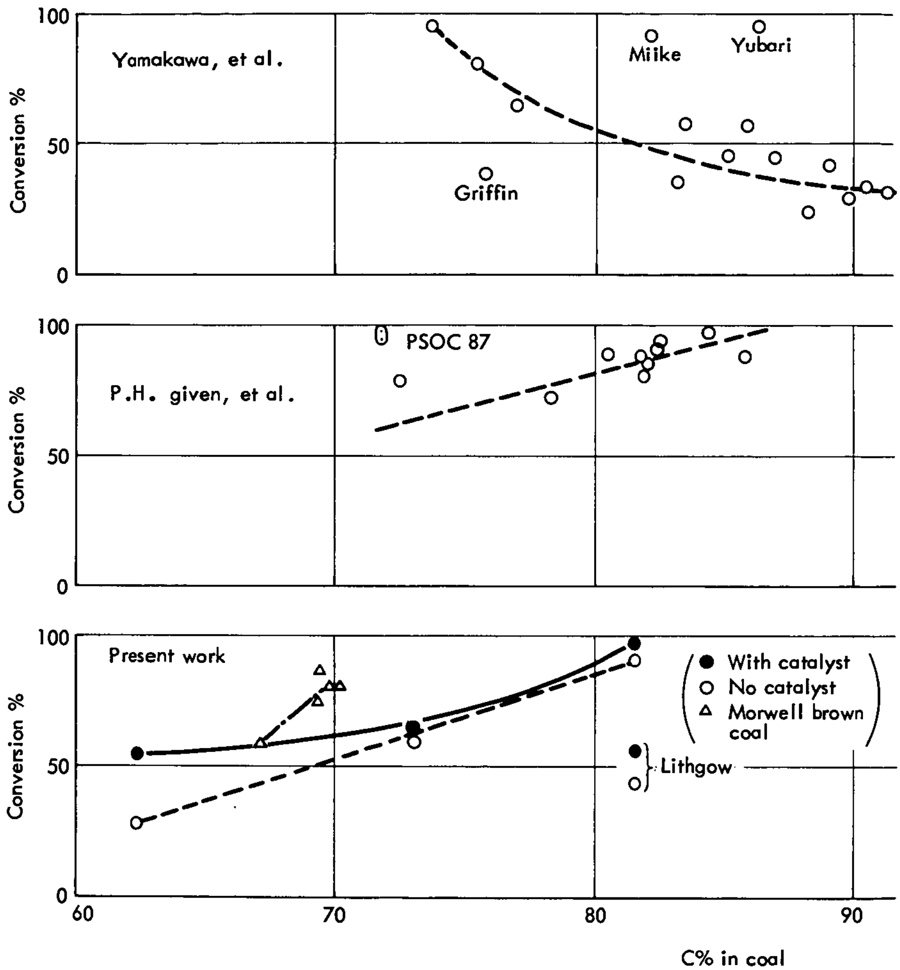

Fig. 2 Relationship between conversion and C% in coal

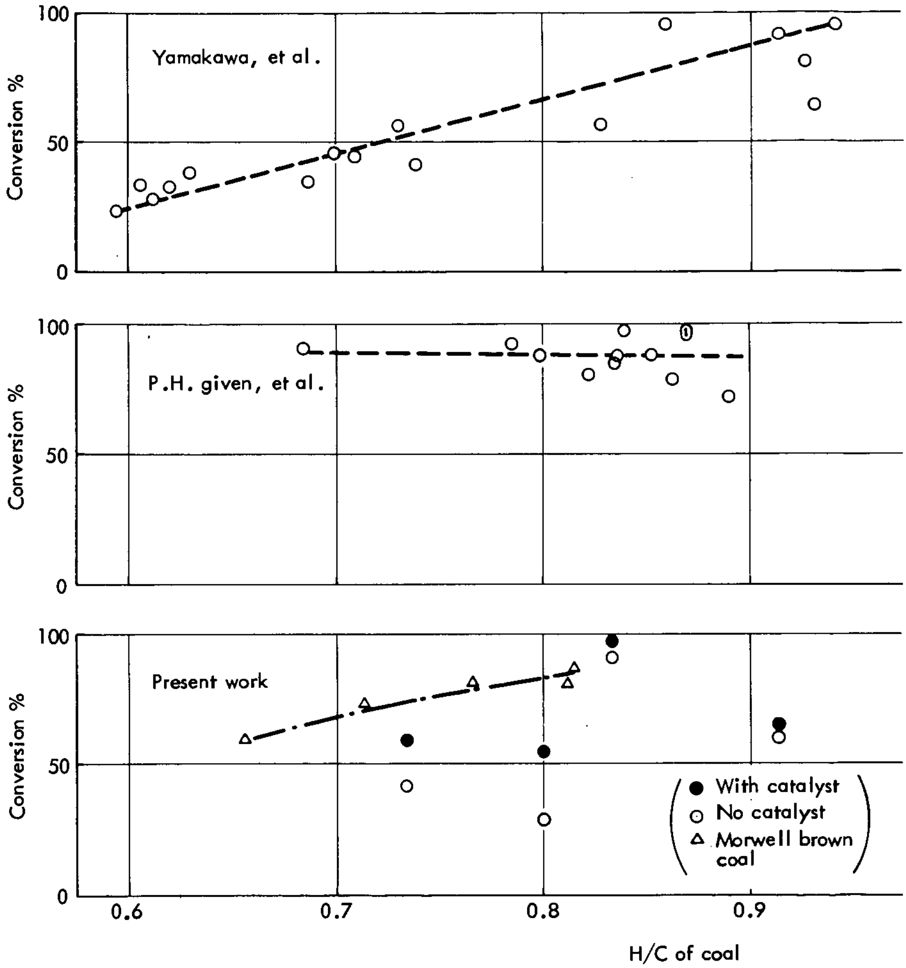

Fig. 3 Relationship between conversion and H/C of coal

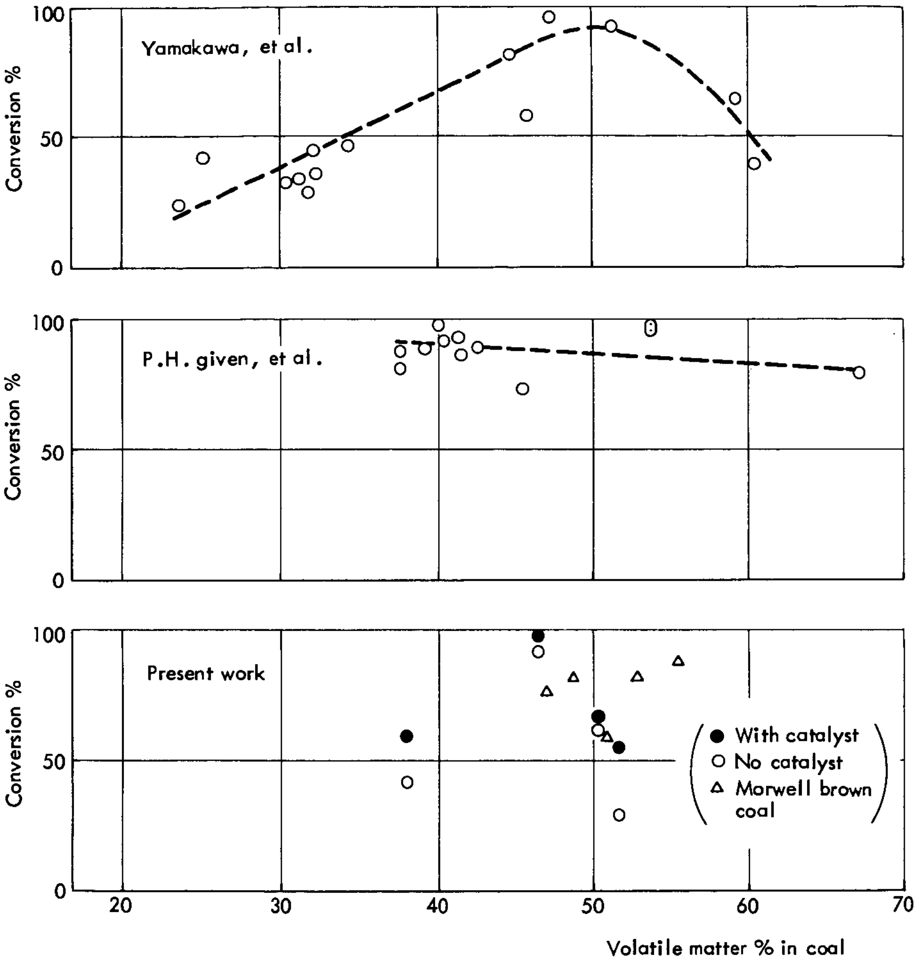

Fig. 4 Relationship between conversion and volatile matter % in coal

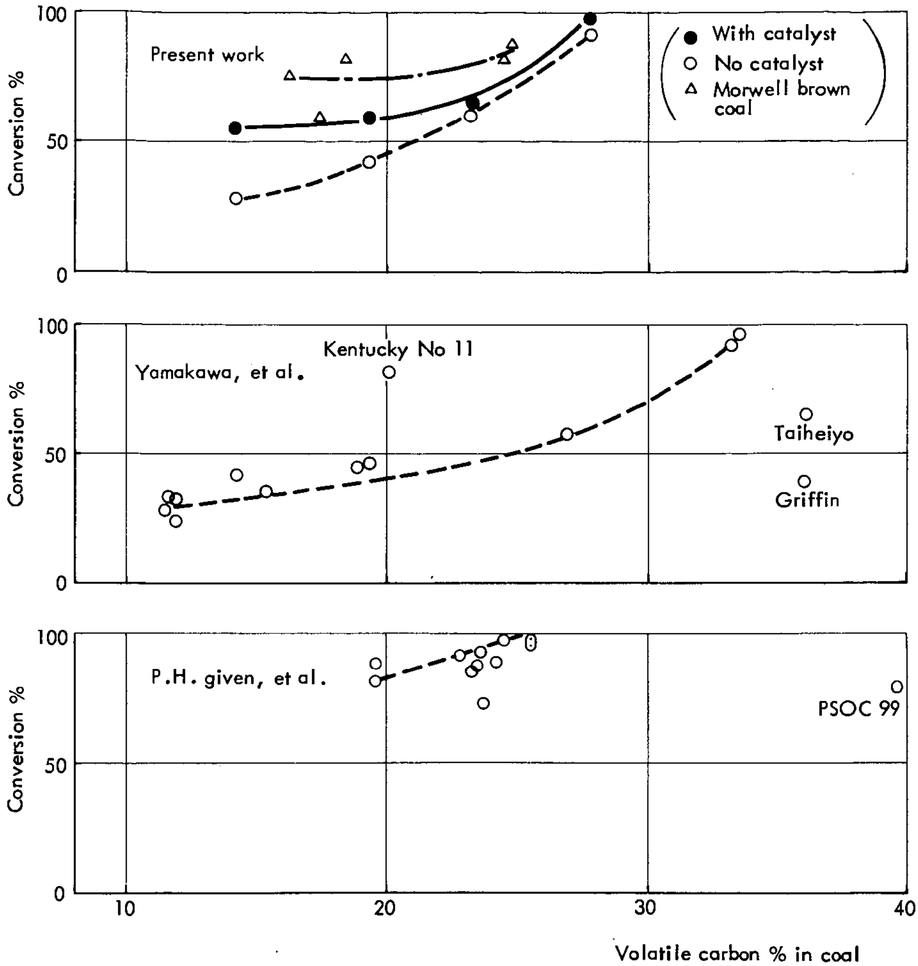

Fig. 5 Relationship between conversion and volatile carbon % in coal

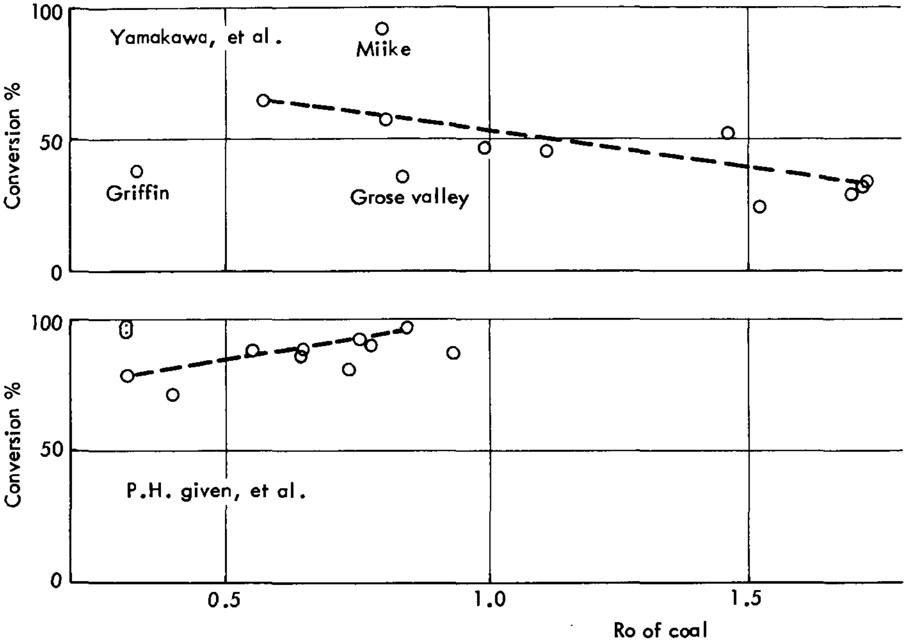

Fig. 6 Relationship between conversion and mean maximum reflectance

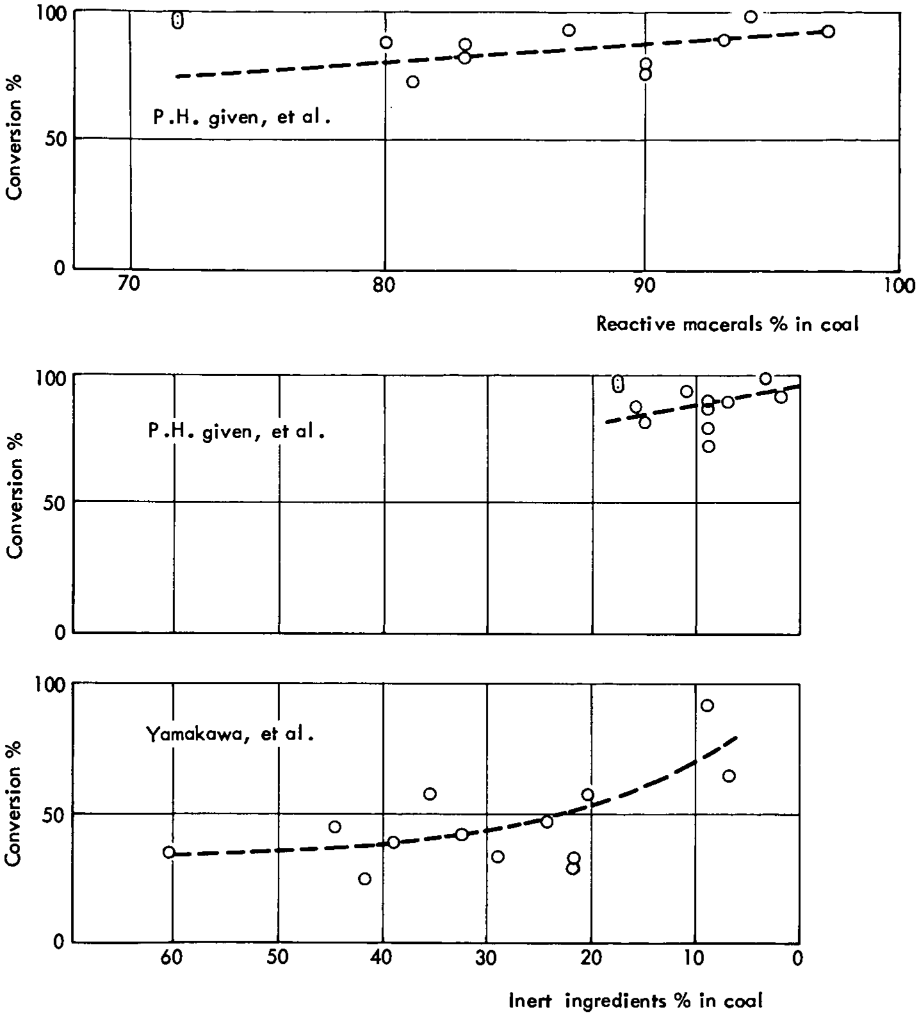

Fig. 7 Relationship between conversion and petrographic components % in coal