The Memoirs of the Geological Society of Japan, No. 15, p. 205~222, Nov. 1978.

INFLUENCE OF TEMPERATURE ON COALIFICATION OF TERTIARY COAL IN JAPAN

Toshio SHIMOYAMA* and Azuma IIJIMA**

Introduction

Japanese coals are of quite wide range of ranks from lignite to anthracite. They occur mostly in the Paleogene measures which are widespread in Japan. The geological feature of the Paleogene measures varies in various coal-fields and so does the rank of coal. Therefore the Japanese coals are ideal for a comparative rank study of coalification.

TANAI (1969), and TANAI and HUZIOKA (1967) summarized the Tertiary floras of Japan and explained that the floristic composition and components in the Paleogene time are of sub-tropical or warm-temperature and have been maintained themselves during the time. Such uniformity of vegetable substances is also favorable for the study of coalification.

SHIMOYAMA and IIJIMA (1974, 1977) examined which factors influence the coalification of Japanese Tertiary coals utilizing the diagenetic zeolite zones associated within coal measures, and they explained it is essentially temperature dependent. In this paper more detailed comparative rank study is presented to determine the thermal history of coalification.

Coalification and Rank Parameters

Coals are derived by metamorphism from geologically buried original plant materials … the process of metamorphism is called coalification. Coals are classified according to their degree of coalification or the degree of maturity into ranks: lignite, subbituminous coal, bituminous coal and anthracite, and based on two major areas of utilization, combustion and carbonization into two categories, non-coking and coking coals.

Among the various elementary parameters relating to physical and chemical properties of coals, the followings are general understandings as to the relationship of rank to constituents of coals. As coalification increases, the moisture, volatile matter and oxygen content correspondingly decrease, while the calorific value and carbon content correspondingly increase. These relation hold true through most stages of coalification or metamorphism but may not be valid through the final stage. For example, the moisture progressively decreases with increasing coalification until the carbon content reaches about 90% and after that it progressively increases (THE COAL MINING RESEARCH CENTRE, 1973). The JIS classifies coals according to the calorific value calculated on mineral matter free basis for the lower rank coal and fuel ratio for the higher rank coal.

For the purpose of genetical classification of the rank of coal, however, the elementary composition measured by ultimate analysis is used. KREVELEN (1950) discussed the reactions that take place during coalification such as decarboxylation, demethanation, oxidation, etc. in the diagram in which the atomic hydrogen to carbon ratio are plotted against the atomic oxygen to carbon ratio. Plot of coals on the diagram is called KREVELEN's coal band. Fig. 1 shows the position of coal bands for Japanese and Euramerican coals (THE COAL MINING RESEARCH CENTRE, JAPAN, 1973). The atomic hydrogen to carbon ratio of the Japanese Tertiary coal is

* Mitsui Mining Co., Ltd. Muromachi 2-1, Nihonbashi, Tokyo, 103 Japan.
** Geological Institute, University of Tokyo, Hongo, Tokyo, 103 Japan

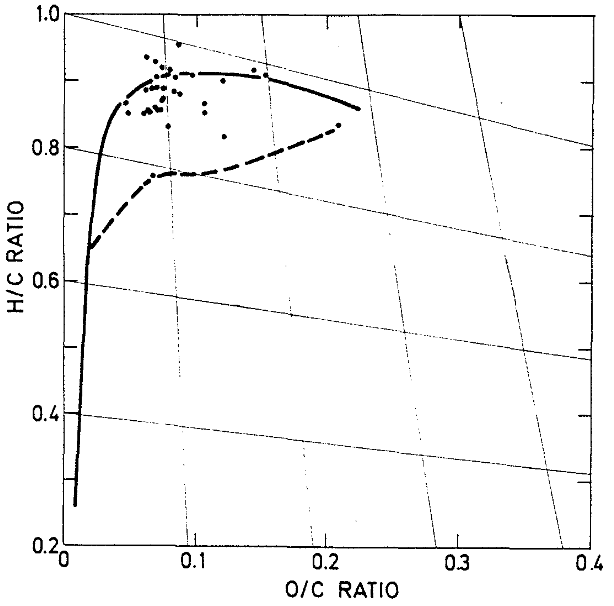


Fig. 1. Diagram showing the KREVELEN's coal band of the Japanese Tertiary coal. Solid and broken curves represent general trends of the Japanese Tertiary and the Euramerican Paleozoic coal respectively.

higher than that of the Euramerican Paleozoic coal of the same range of the oxygen to carbon ratio (FUJII *et al.*, 1966).

The petrographic composition of coal is expressed in terms of macerals and microlithotypes. Reflectance of macerals particularly on one component, vitrinite, is used as the precise method to determine the rank of coal since as the rank of coal increases so does the reflectance.

Since SHAPIRO and GRAY (1964) discussed the predicting method for the coke strength, reflectance analysis (petrographic analysis) has been widely utilized as an important technique in the coke manufacturing to evaluate coking characteristic and its blendability.

The reflectance of coal increases in inverse proportion to the atomic hydrogen to carbon ratio below 0.8% of the hydrogen to carbon ratio, while it decreases, gradually in case of the Japanese and rapidly in case of the foreign coals,

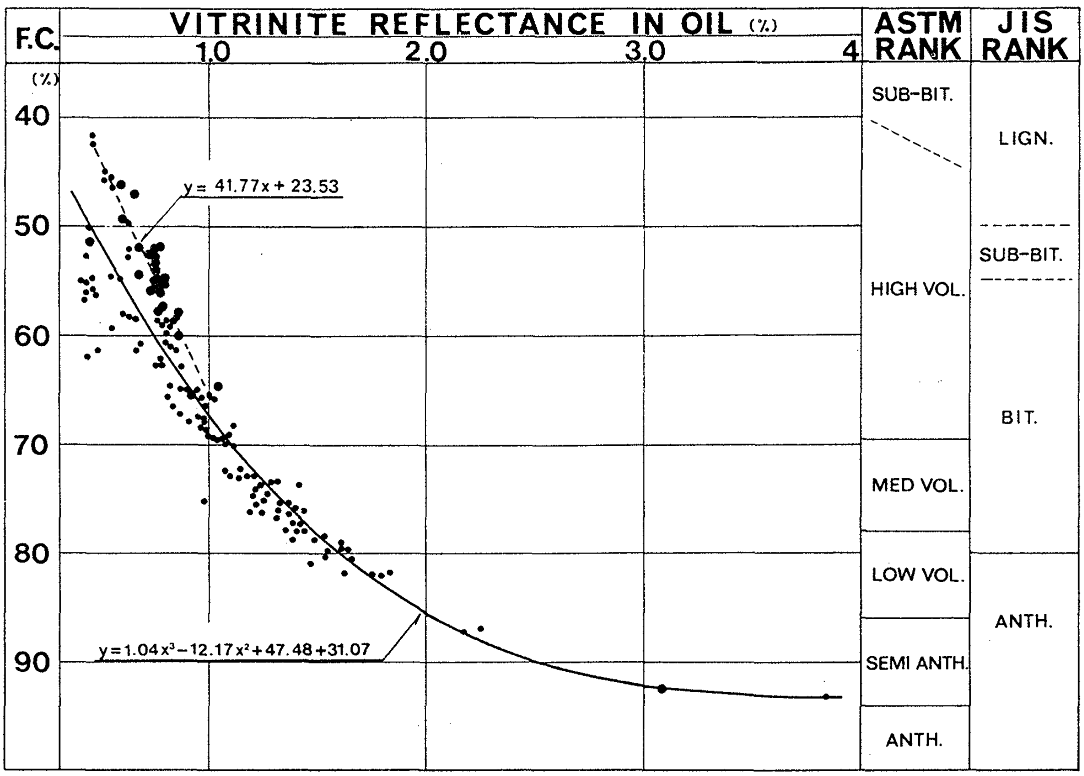


Fig. 2. Diagram showing correlation between the reflectance on vitrinite component and the fixed carbon of the whole coal.

with the increase of the hydrogen to carbon ratio above 0.8% of the hydrogen to carbon ratio (SUGIMURA *et al.*, 1966). FUJII *et al.* (1966) noted that the hydrogen to carbon ratio of the Japanese Tertiary coal is higher than that of the Euramerican Paleozoic coal of the same range of the hydrogen to carbon ratio.

Fig. 2 illustrates the correlation between the reflectance on vitrinite component and the fixed carbon of the whole coal. This figure is established based on 135 samples: 16 American, 23 Australian, 64 Canadian, and 32 other countries which were analyzed in our laboratory, which is shown by the solid line.

The broken line represents the correlation for the Japanese Tertiary coal which is of more

Table 1. The typical analysis of the Japanese coal from the major coalfields.

Locality	Formation	Ro	H/C	O/C	VM	FC	Cal/gr
Tempoku*	Soya	0.45	0.836	0.208	48.3	51.7	5,780
Chikubetsu*	Haboro	0.53	0.909	0.152	50.5	49.5	6,890
Joban*	Iwaki	0.55	0.900	0.120	52.7	47.3	7,400
Kushiro*	Harutori	0.59	0.917	0.143	53.9	46.1	7,120
Bibai (Mitsui)	Bibai	0.57	0.909	0.097	45.7	54.3	8,040
Tagawa	Sanjaku-Goshaku		0.866	0.105	51.1	48.9	8,290
Takamatsu*	Onga	0.72	0.817	0.121	44.1	55.9	7,310
Tempoku (MITI-Wakkanai)		0.72	0.891	0.059	44.2	55.8	8,450
" " "		0.76	0.826	0.056	43.2	56.8	8,400
Bibai (Mitsubishi)	Bibai	0.75	0.852	0.106	48.2	51.8	8,120
Akabira*	"	0.78	0.888	0.074	44.8	55.2	8,090
Sunagawa (Mitsui)							
" "	Bibai	0.76	0.884	0.082	45.3	54.7	8,220
" "	"	0.78	0.858	0.072	43.1	56.9	8,120
" "	"	0.76	0.831	0.078	41.9	58.2	8,210
" "	Wakanabe	0.82	0.874	0.074	42.9	57.1	8,300
" "	"	0.86	0.858	0.072	40.0	60.0	8,250
" "	Yubari	0.83	0.874	0.073	41.6	58.4	8,280
" "	Noborikawa	0.92	0.757	0.066	34.2	65.7	8,350
" "	"	0.75	0.852	0.048	44.6	55.4	8,380
" "	"	0.77	0.853	0.064	42.2	57.8	8,450
Ashibetsu (Mitsui)							
" N3 -	Bibai	0.76	0.954	0.086	44.8	55.2	8,270
" N3 - 85	"	0.79	0.881	0.087	44.0	56.0	8,170
" N1 - 200	"	0.82	0.860	0.068	43.5	56.5	8,370
" N1 - 300	"	0.85	0.859	0.069	42.1	57.9	8,330
" N1 - 410	"	0.85	0.850	0.060	41.8	58.2	8,410
" N1 - 410	"	0.74	0.919	0.073	45.0	55.0	8,200
" N3 - 0	Yubari	0.75	0.905	0.083	47.5	52.0	8,260
" N3 - 85	"	0.74	0.916	0.079	46.6	53.4	8,330
" N1 - 200	"	0.79	0.890	0.069	45.2	54.3	8,450
" N1 - 300	"	0.77	0.905	0.069	48.2	51.8	8,400
" N1 - 400	"	0.79	0.856	0.063	44.9	55.1	8,320
" N5 - 505	"	0.76	0.929	0.068	44.9	55.1	8,380
Miike							
"	Kattachi	0.67	0.935	0.062	50.5	49.5	8,530
"	Nanaura	0.72	0.886	0.063	47.8	52.2	8,600
"	Toka	0.75	0.889	0.065	46.1	53.9	8,530
Yubari*	Yubari	0.89	0.866	0.046	41.6	58.4	8,520
Takashima	Hashima	1.03	0.846	0.049	38.6	61.4	8,630

* The Coal Mining Research Centre, Japan (1973).

than 35% fixed carbon content, which is established based on 33 samples. The reflectance of the Japanese coal of the above mentioned range of fixed carbon content is higher than that of the foreign coal of the same range of fixed carbon content. For the coal which contains less than 35% fixed carbon content the reflectance does not differ from each other between the Japanese and foreign coals. The correlation is good although there is considerable scatter of values particularly on relatively lower rank coal. The simple correlation(R) calculated for the Japanese Tertiary coal is 0.820 and the multiple correlation for the foreign coal is 0.947. The reflectance was estimated using this diagram when it was not available.

The typical analysis of the Japanese coal from the major coal fields and some of the data used for the above calculation are given in Table 1. The Tempoku and Chikubetsu coalfields yield the Miocene lignite and subbituminous coal, respectively. The Joban, Kushiro and Mitsui Bibai coalfields yield the Paleogene subbituminous coal. The rest are of the Paleogene coking bituminous coal. The critical reflectance to separate the coking bituminous coal from the non-coking coal is about 0.6. The critical oxygen to carbon ratio to distinguish the coking from the non-coking is about 0.120 with a range of the atomic hydrogen to carbon ratio from 0.650 to 0.900.

Representative Coalfields and Regional Rank of Coal

Palaeogene coal bearing strata are widespread in the Japanese Islands except in the Setogawa-Nakamura geosyncline on the Pacific side of Southwest Japan (Fig. 3).

The Paleogene coal measures consist of thick piles of marine and fresh water sediments. The geological correlation of the main coalfields is shown in Table 2.

The Paleogene coal measures in Hokkaido are represented by the Ishikari Group which is fully developed in the Ishikari coalfield where it attains a maximum thickness of 4,000m. The Ishikari coalfield extends about 85 km north-south, 30 km east-west and is devided into two districts, Sorachi in the north and Yubari in the south. The schematic east-west geological profiles of both districts reveal that the coal bearing formation have been affected by intense faulting and folding, particularly in Yubari district (Fig. 4). The original depth of burial of the Ishikari group is estimated about 5,000 to 6,000 m. Except for the non-coking subbituminous coal in the Mitsui-Bibai area of the Sorachi district, the coal interbedded with the Bibai, Wakanabe, Yubari and Noborikawa Formations of the middle and lower Ishikari Group is coking bituminous. The coal in the upper Ishikari Group is subbituminous.

The Paleogene coal measures in eastern Hokkaido are represented by the Urahoro Group which is 2,000m thick at the most. The eastern part of the Kushiro coalfield, namely the Harutori area shows gentle homoclinic structure offshore, while the western part shows a folding structure. The maximum depth of burial of the Urahoro Group is estimated about 2,500m in the Harutori area. The coal interbedded with the Urahoro Group is subbituminous.

The Paleogene sediments in the Joban coalfield in Northeast Honshu are only 200~700 m thick. Coal seams are interbedded with the Oligocene Iwaki Formation of the Shiramizu Group which is uncomformably overlain by the Neogene sediments being about 1,000m thick, but it is unlikely that the total depth of burial exceeds 2,000m. The schematic east-west geological profile illustrates that the strata are gently inclined and thickenning offshore (Fig. 4). The rank of the Joban coal is low from lower rank subbituminous to lignite.

The Kuji coalfield in Northeast Honshu shows the coal occurrences similar to the Joban coalfield.

The Paleogene sediments in northern Kyushu are distributed in several separate coal basins.

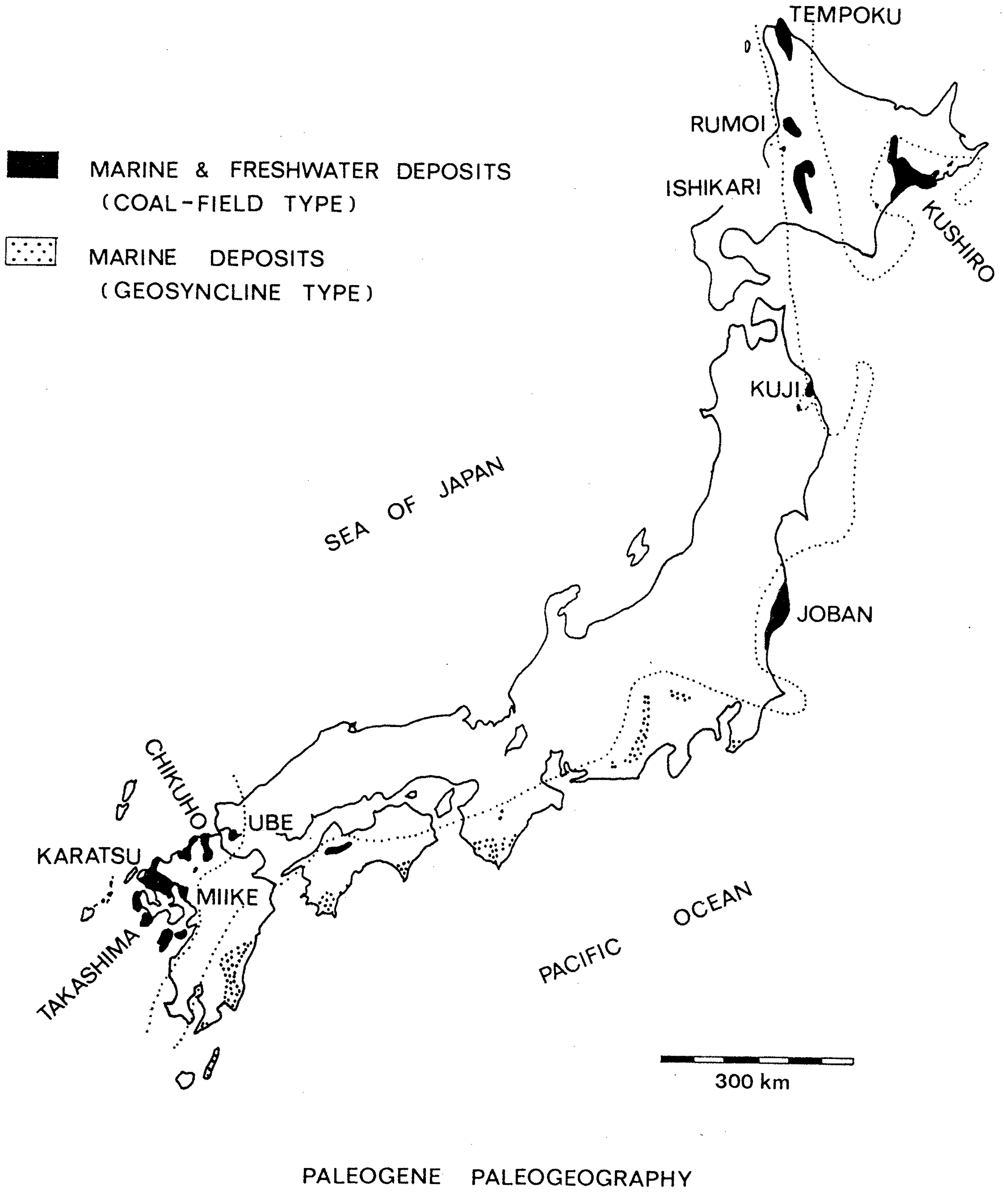
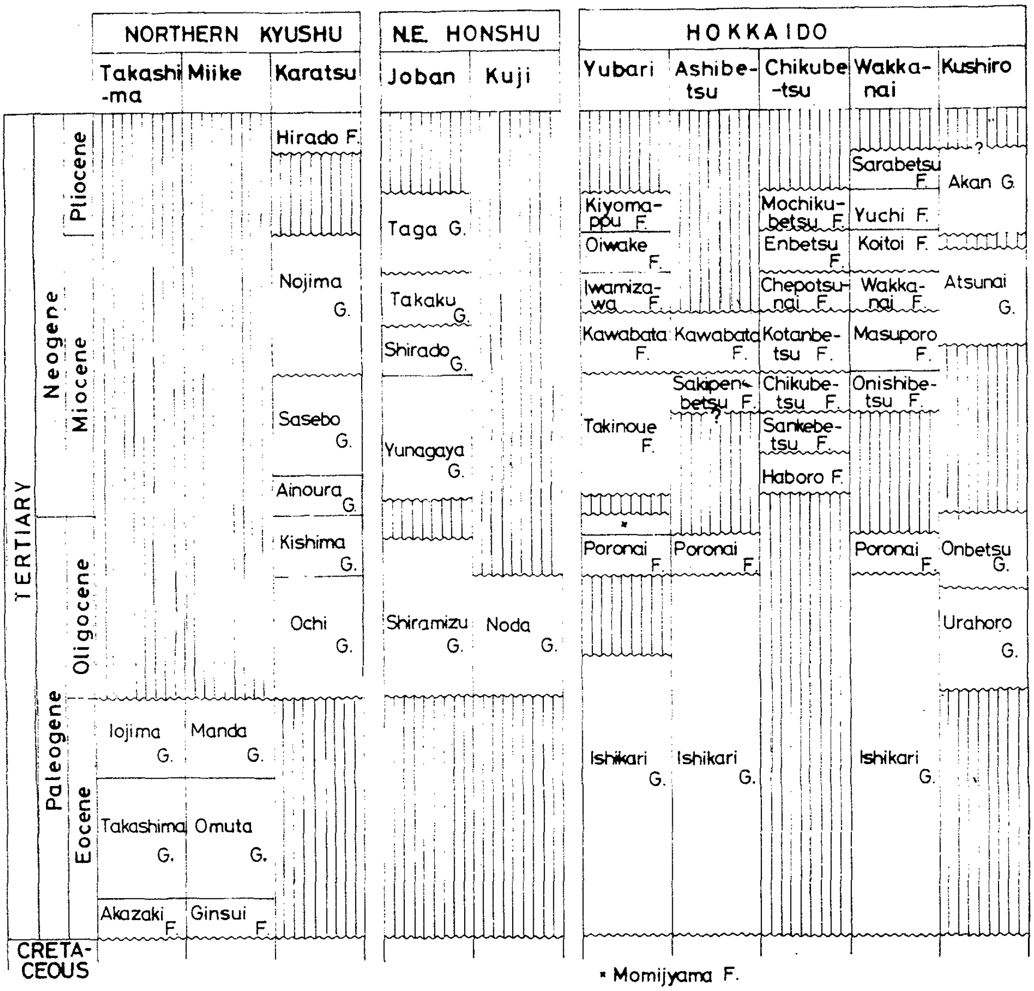


Fig. 3. Map showing the Paleogene Paleogeography of the Japanese Islands.

Table 2. The geological correlation of the main coalfields in Japan.



They thicken toward northwest and west which attain the maximum thickness of 2,000m.

The Miike coalfield lies alongshore and beneath the Ariake-Bay, where the Miike colliery — the largest coal mine in Japan — located, whose annual production is 6 million tons. The productive coal seams in the Miike colliery occurs in the Eocene Omuta and Manda Groups. The total thickness of the Paleogene sediments is only 500m thick. The schematic east-west geological profile illustrates that the strata are gently inclined offshore. The maximum depth of burial probably obtained 2,000m when overlain by the Neogene sediments (Fig. 4). The Miike coal is coking bituminous.

The Takashima coalfield which lies outside of the Nagasaki Port produces quite similar but a little higher rank bituminous coal than the Miike coalfield. The coalfield shows similar geological history to the Miike coalfield.

In the Karatsu coalfield the Paleogene sediments are more than 1,000m thick and are overlain by the Neogene sediments which are at least 3,500m thick. The coal seams are interbedded with the Oligocene Ochi and Miocene Sasebo Groups. The field is slightly deformed. The rank of coal of both the Ochi and Sasebo Groups are coking bituminous. The depth of

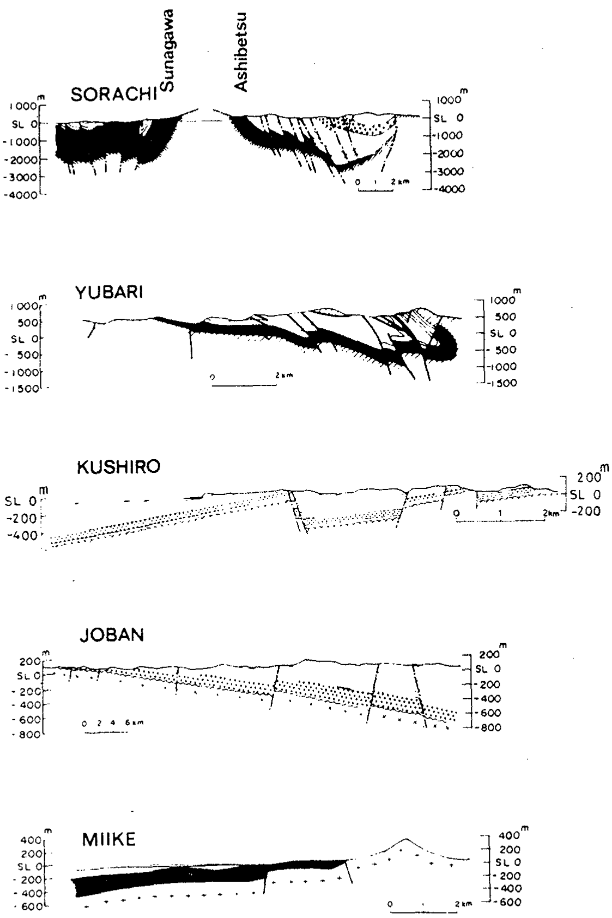


Fig. 4. Geological profiles of representative Paleogene coalfields of Japan.

burial is estimated 3,000m and 5,000 for the Sasebo and Ochi Groups respectively as measured from the stratigraphical succession.

Geologic Significance in Rank of Coal – Effect of Depth of Burial and Tectonic Deformation

From aforementioned description it is difficult to find any relation between the rank of coal and the total depth of burial. This is illustrated in Fig. 5. In the Miike, Takashima and Joban coalfields where the maximum depth of burial is estimated as 2,000m, the rank of coal expressed in terms of reflectance in the former two coalfields ranges from 0.8 to 1.03, whereas in the latter the rank is 0.55. In the Ishikari coalfield where the depth of burial is estimated as 5,000~6,000m, the rank ranges from 0.78 to 0.89, whereas in the Miike and Takashima notwithstanding shallower depth of burial, the rank is similar to the Ishikari.

The degree of deformation is much greater in the Ishikari coalfield than in the Miike. It has not, however, been observed at the middle and lower part of the Ishikari coalfield that tectonic deformation may increase their ranks compared with the Miike. The upper part of the Ishikari Group yields subbituminous coal notwithstanding the great tectonic deformation.

These facts strongly suggest that coalification is not essentially affected either by the depth of burial (confining pressure) or by the degree of tectonic deformation. It can be accepted that the essential factor of coalification is neither confining pressure nor tectonic pressure.

Diagenetic Zeolite Zones in the Tertiary Coalfields

Since IIJIMA (1961) first reported the occurrence of clinoptilolite from the Harutori colliery in the Kushiro coalfield, five species of authigenic zeolites have been found in coal measures and their underlying and overlying strata in most of the Japanese Paleogene coalfields (SHIMOYAMA and IIJIMA, 1974, 1977). They are clinoptilolite, mordenite, analcime, heulandite and laumontite. They occur exclusively, except for some laumontite, as an alteration product of silicic glass in vitric tuffs which are sparsely interbedded with coal seams, shales and sandstone as persistent layers usually 10~100cm thick. No distinction in the mode of occurrence of zeolites was recognized among freshwater, brackish and marine deposits. Clinoptilolite and mordenite are altered from silicic glass, and are associated usually with montmorillonite and low cristobalite. Analcime replaces the precursor clinoptilolite and mordenite, being associated with chlorite, montmorillonite-chlorite mixed layer and quartz. Relics of clinoptilolite and mordenite are not un-

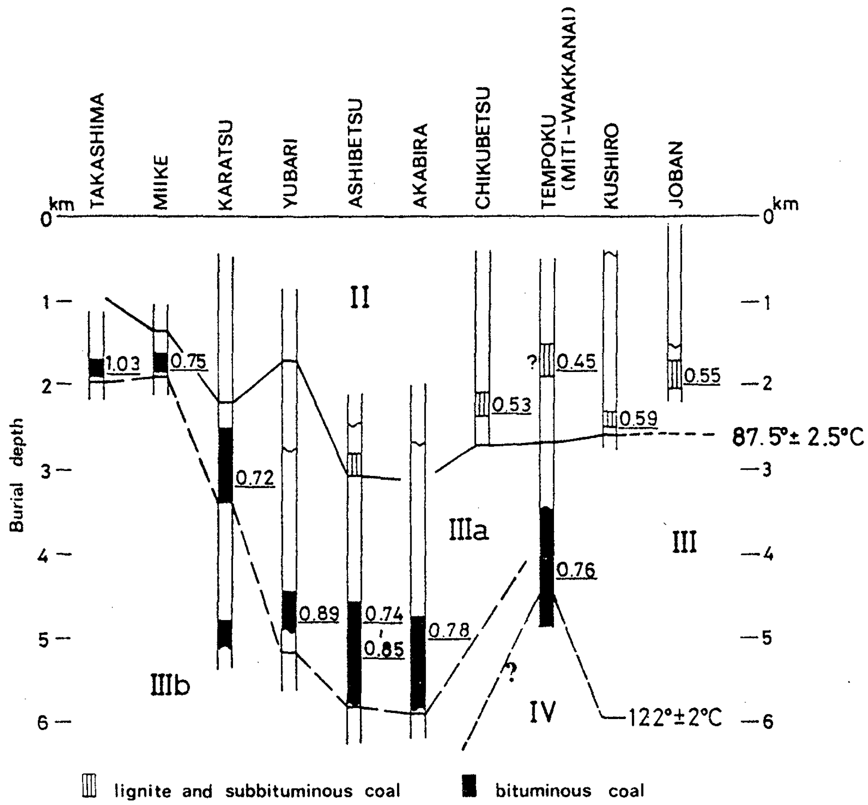


Fig. 5. Relationship among coal ranks, zeolite zones and depth of burial in the Tertiary coalfields in Japan.

common in analcimic tuffs. Heulandite changes from the precursor clinoptilolite, and is associated commonly with analcime, chlorite, montmorillonite-chlorite mixed layer and quartz. Laumontite is associated usually with chlorite, montmorillonite-chlorite mixed layer, quartz and frequently with calcite and analcime. It replaces the precursor heulandite and montmorillonitic clay in silicic tuffs. Also it occurs in the cement of coarser grained tuffaceous or epiclastic sandstone in which it frequently replaces plagioclase. These authigenic zeolites found in Japanese Paleogene coalfields are distributed systematically in a vertical zonal arrangement within a limited local stratigraphic succession, as shown in Fig. 5. Clinoptilolite and mordenite always appear in the shallowest zone. Analcime characterizes the zone underlying the clinoptilolite-mordenite zone. Heulandite and Laumontite occur locally in the upper and lower parts, respectively, of the analcime zone. Albite zone distributes under the analcime zone.

Many researches of sedimentary zeolites in Japan have revealed (IIJIMA and UTADA, 1971, 1972; IIJIMA, 1975, 1978) that alkali zeolitic reactions are substantially progressed with increase of temperature during the burial diagenesis of marine and freshwater sediments, so that the zeolites are progressively distributed in a vertical zonal arrangement. Two zeolitic reaction series progress during the burial diagenesis in silicic tuffs and tuffaceous sediments interbedded with the Tertiary and Cretaceous strata in the Japanese oilfield:

(1) Alkali zeolite reaction series;
Silicic glass + H_2O − clinoptilolite and/or mordenite − analcime + quartz − albite + quartz.

(2) Calcic zeolite reaction series;
Clinoptilolite + low cristobalite − heulandite

+ quartz − laumontite + quartz − prehnite + quartz.

The former series occur far more regionally than the latter. Hence, based on the alkali zeolite reaction series, four diagenetic zones are defined as follows (Fig. 6):

Zone I characterized by the presence of unaltered silicic glass
Zone II characterized by the presence of clinoptilolite and/or mordenite
Zone III characterized by the presence of analcime
IIIa characterized by heulandite-analcime assemblage
IIIb characterized by laumontite-analcime assemblage
Zone IV characterized by the presence of albite replacing analcime

The depth of burial and the temperature increase from Zone I to Zone IV in the same area. It is significant that the zeolite zoning in the coalfields aforementioned is essentially the same as the zoning in the oilfield.

In the Ishikari coalfield and in the MITI Wakkanai drill cores in Tempoku, the above four zones were identified. In the Ashibetsu colliery in the Sorachi district of the Ishikari coalfield, analcime is noted in the lower part of the Yubari Formation, which includes coal seams of rank 0.78. In the MITI Wakkanai drill cores analcime is noted at 1,179.2 m. Plagioclase in sandstones changes extensively to albite at a depth below 3,275.9 m which belong to Zone IIIb or IV. The rank of coal penetrated at 3,370 and 3,452 m is 0.72 and 0.76 respectively. In the Harutori area of the Kushiro coalfield and the Kuji and Joban coalfields only Zone II is identified in which the rank of coal is low, 0.53∼0.59. In the Miike, Takashima and Karatsu coalfields, Zones II, IIIa and IIIb are identified. The Miike productive coal seams of which ranks are somewhere around 0.80 are included in Zone IIIa. As can be seen in Fig. 5 the specific correlation of the rank of coals with the zeolite zones is recognized throughout the Japanese Tertiary coalfields. All of the lignite and subbituminous coal, on the one hand, belong to Zone II for example in the Kushiro, Kuji and Joban coalfields. The coking bituminous coal, on the other hand, belongs exclusively to the Zone III like in the Ishikari, Miike, Takashima and Karatsu coalfields.

Thermal History Based on Zeolite Zones

IIJIMA and UTADA (1971) examined the most typical present-day burial zeolitic diagenesis in the Niigata oilfield on the coast of Sea of Japan and explicitly concluded that the alkali ze-

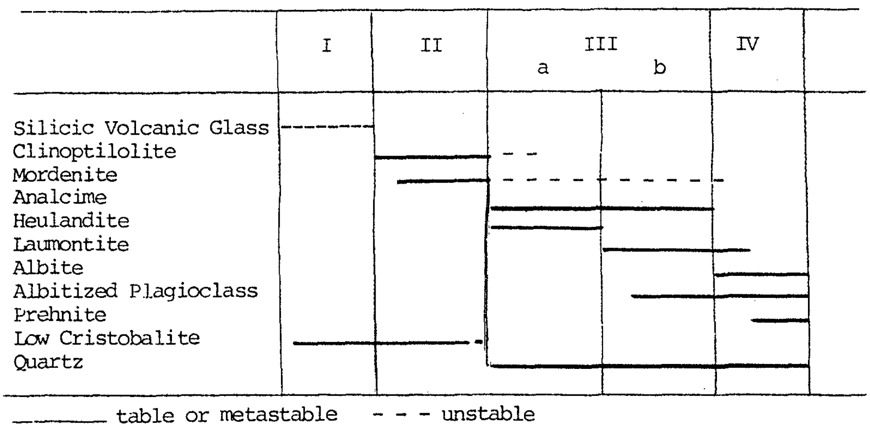


Fig. 6. Fundamental zoning due to burial zeolitic diagenesis of cilicic vitric tuffs in Japan.

olite reactions are essentially not P_{H_2O} dependent but temperature dependent. This conclusion is further confirmed by the petrological analyses of deep cores from six MITI drillholes in central Hokkaido (IIJIMA, 1975, 1978a, b). The temperature and depth at which the zeolite zone boundary occurs in ten holes are tabulated in Table 3. In five holes on the upper side of the table, the present-day depth is geologically interpreted to represent the maximum depth of burial and progress the burial diagenesis. Therefore, the temperature and depth indicate the starting point-equilibrium-of the alkali zeolite reactions. The temperature is 84° ~ 91 °C, averaging 87°C at the top of Zone III (analcime zone), and is 120° ~ 124°C, averaging 122°C at the top of Zone IV (albite zone). These values are very good convergence considering from the error of bottomhole temperature measurements. On the contrary, the depth is quite variable— that means variable P_{H_2O}. In five holes on the lower side of the table, the temperature at the top of both Zone III and IV is much lower than in the upper five holes. The overburden of approximately 1,000 ~ 1,500m thick is geologically interpreted to have been removed by uplift and erosion in the lower five holes, so that the present temperature should be lower than the original temperature at which the reactions started. Therefore, it is concluded that the alkali zeolite reactions are temperature dependent at a depth range of at least less than 5km.

Experimental studies seem to support the above conclusion that the zeolite zones specified by the alkali zeolite reactions are temperature dependent. FYFE *et al.* (1958), COOMBS *et al.* (1958), and NAKAJIMA (1973) studied the stability fields of analcime-albite reaction on the basis of hydrothermal synthesis. CAMPBELL and FYFE (1965) discussed the analcime-albite equilibria and THOMPSON (1971) revised it on the basis of the reaction-rate method. These results are not al-

Table 3. Temperature and depth at which the zeolite zone boundary occur in the MITI drill holes.

Drill	II - III		III - IV		Remarks
	T°C	depth m	T°C	depth m	
MITI Obuchi*	84	2,900	120	4,160	The present-day depth is geologically interpreted to represent the maximum depth of burial and progress the zeolite burial diagenesis.
MITI Shimoigarashi*	87	3,500	124	4,480	
MITI Masugata*	91	3,490	124	4,500	
Yabase oilfield**	88	1,700	120	2,500	
MITI Hamayuchi	85	3,250	120	4,500	
MITI Wakkanai	60	2,000	85	3,250	The overburden of approximately 1,000 - 1,500 m thick is geologically interpreted to have been removed by uplift and erosion.
MITI Enbetsu	55	1,350	95	3,750	
MITI Nanporo	55	2,200	-	-	
MITI Karumai	50	1,750	90	4,020	
MITI Niikappu	50	2,600	-	-	

* Iijima and Utada (1971) ** Huzioka and Yoshikawa (1969)

ways coincident. However all results represent that the reaction is essentially temperature dependent below 2kb P_{H_2O}. This is not inconsistent with the above-stated field evidence, because P_{H_2O} is usually less than 2kb in zeolitic burial diagenesis. The difference in the equilibrium temperatures in between the field and the laboratories is caused principally by the concentration of Na^+ in interstitial water which acts to lower the reaction temperature (IIJIMA, 1975, 1978a, b).

As mentioned before, no distinction in the mode of occurrence of zeolites was recognized among freshwater, brackish and marine deposits in the coal fields. According to the study of carbonate diagenesis in the Paleogene coalfields by MATSUMOTO (1978), intense mixing of interstitial water of such deposits occured at a depth of burial of 300 ~ 500 m ; and the fresh water sandstones of the Ikushunbetsu Formation in the Yubari district contain the brine water same as in the marine Wakanabe Formation. It is, therefore, reasonable to consider that the zeolite zoning in the coalfields is essentially temperature dependent and to deduce the same reaction temperature as that in the oilfields.

It could, therefore, be concluded that the formation temperature of the non-coking coal, which associated with Zone II, is as much as about 85 °C, while formation temperature of coking coal which associated with Zone III range from 85° to 125°C.

Main Factor to Control Coalification

The factors which influence coalification are discussed by many researchers, KARWEIL (1956), KUYL and PATIJN (1963), TEICHMÜLLER and TEICHMÜLLER (1966, 1969), KISCH (1969, 1974), HACQUEBARD and DONALDSON (1970, 1974), BOSTICK (1974), CASAÑO and SPARKS (1974). Most of them generally agree that the most important factors are temperature and to less time. And also time is believed to have little importance if the temperature was low and to have much importance if heating was severe (TEICHMÜLLER and TEICHMÜLLER, 1968). If coalification took place in the pre-orogenic stage, the rank should change with the stratigraphic position, because the temperature at depth is related to the original depth of burial and geothermal gradient.

Changing the rank of coal with the stratigraphic position at the Mitsui Sunagawa and Mi-

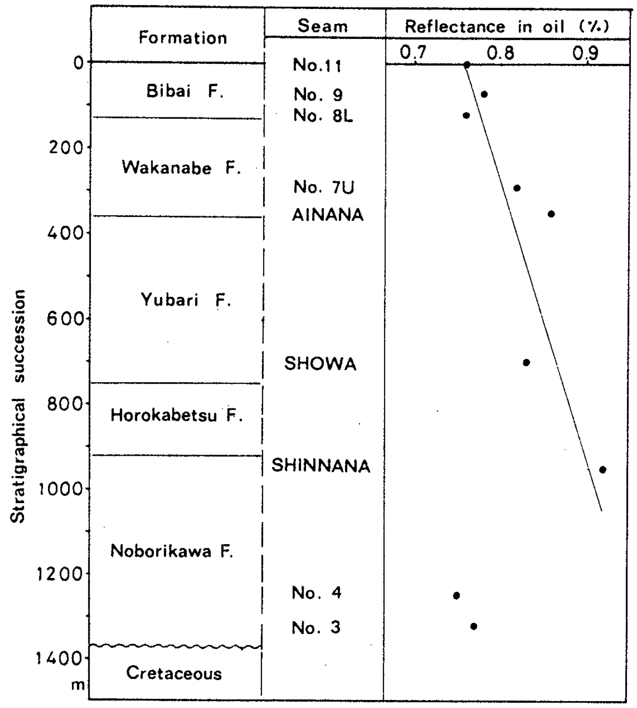


Fig. 7. Changing the rank of coal with the stratigraphic position at the Mitsui Sunagawa colliery.

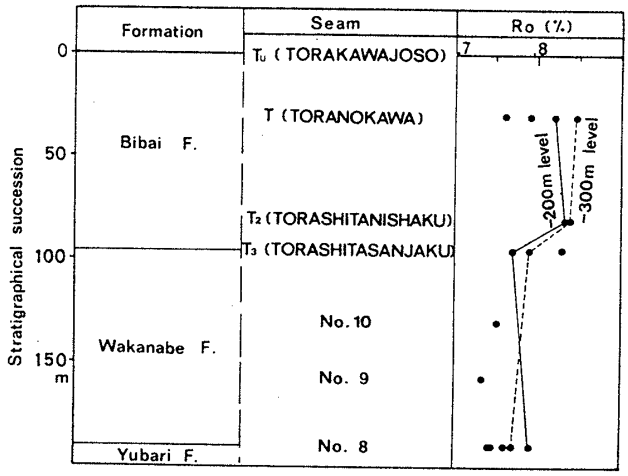


Fig. 8. Changing the rank of coal with the stratigraphic position at the Mitsui Ashibetsu colliery.

tsui Ashibetsu collieries in the Sorachi district, Ishikari, is shown in Fig. 7 and 8 (see Fig. 4). At first glance of the Mitsui Sunagawa diagram, although there is scatter in data, the rank of coal seems to increase gradually as descending stratigraphic succession except the lowermost position in the lower part of the Noborikawa Formation, where the rank of coal shifts to low. This suggest that the rank of coal in the Sunagawa area would be of pre-orogenic origin, but for the low rank of the lower Noborikawa Formation. In the second place, at the Mitsui Ashibetsu diagram, the rank of coal does not increase alongwith the stratigraphic position. Fig. 9 and 10 show the geologic profiles of the cross-cuts in the Sunagawa and Ashibetsu collieries where samples were taken. The reflectance on the samples are given in Table 1. As can be seen in Fig. 9, though the Yubari and Noborikawa Formations are repeated by several faults, the rank of coal seems to be equivalent to the stratigraphic position except for the lower part of the Noborikawa Formation. Among many faults the Sunagawa fault is a distinctive hinge fault. If, therefore, at the initial depth that existed before faulting, the rank of coal of the lower part of the Noborikawa Formation was lower than in any of other sections as assumed by AIHARA (1977), the movement of the Sunagawa fault which raised the hanging wall to the foot wall may to some extent account for the shifts of the rank of coal to low. However, in the Ashibetsu area there exists no fault, particularly within the section between the Bibai and Yubari formations at −200 m level cross cut, and although the Toranokawa seam is located stratigraphically about 160 m above No. 8 seam, the Toranokawa seam shows rather high reflectance

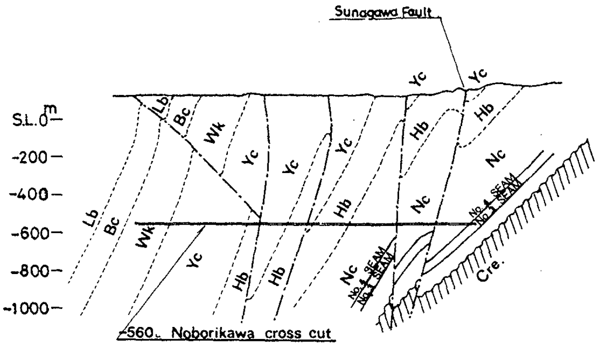


Fig. 9. Geologic profile at −560 level cross cut in the Mitsui Sunagawa colliery.

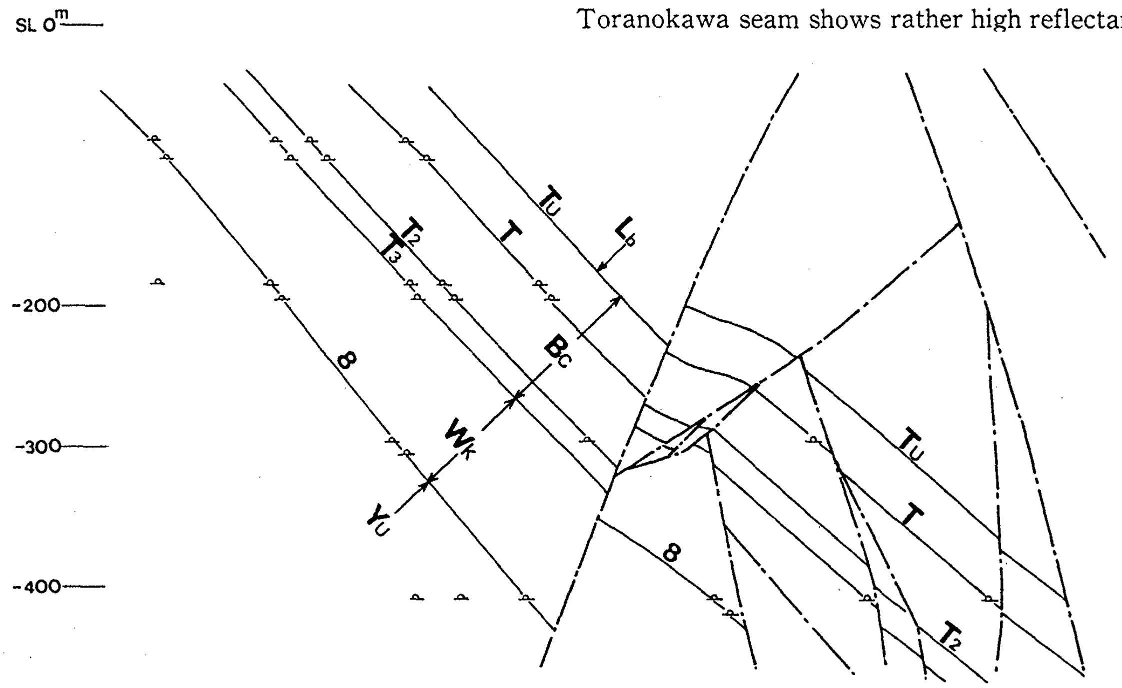


Fig. 10. Geologic profile at N-1 panel cross cut in the Mitsui Ashibetsu colliery.

than No. 8. It is difficult to explain less difference in the rank of coal in the lower horizon than coal in the upper horizon. Such irregularities in the rank alongwith the stratigraphic position was studied in the Saar Basin and explained that thick sandstone beds caused small coalification because in the sandstone the heat may flow so rapidly that the coal rank hardly increases at all with depth (DAMBERGER, 1968). Here, in the Ashibetsu area, the sandstone ratio of the Bibai Formation is 47%, and those of the Wakanabe and Yubari Formations are about 70% and 45 respectively. No. 10 and No. 9 seams which are interbedded with the sandstone predominant Wakanabe Formation show low reflectance. This appears to show that the lithology of the host rock provides the different temperature gradient so that it affects the degree of coalification even within limited area. Analogy may exist in the Sunagawa area, in the lower part of the Noborikawa Formation higher percentage of relatively thick sandstone is present, of which sandstone ratio is about 70%, than in any of other sections of which sandstone ratio is about 40%. This may account for the greater shifts of the rank of coal to low in the lower part of the Noborikawa Formation, though it is subject to debate. The previous discussion, anyhow, has shown that the irregularities in the rank of coal with the stratigraphic position are recognized even in the limited area. Therefore care must be taken to inspect the relationship between the rank of coal and the stratigraphic position in the Sunagawa area.

Fig. 11 shows changes in the rank of coal of No. 8 and Toranokawa seams in the Ashibetsu area. In the Fig., within individual seams, the rank of coal does not change with an increase of the depth of mining ranging from 0 to −1,000m. This concludes that it is likely that the post-deformational depth of burial has had little effect on the rank of coal so that the retrogressive coalification is not apparent. It is geologically interpreted that the maximum burial stage was reached sometime in Miocene, and the faulting was completed before the deposition of the Takikawa Formation of early Pliocene in the Ishikari coalfield. Since duration of burial, therefore, is estimated that it was brief, it is much emphasized that the temperature is main factor to control coalification, which was provided at the depth when buried.

Prediction of Thermal History

Various calculations of the paleo-temperature from the rank of coal have been discussed by many researchers (HUCH and KARWEIL, 1955; KARWEIL, 1956; HACQUEBARD and DONALDSON,

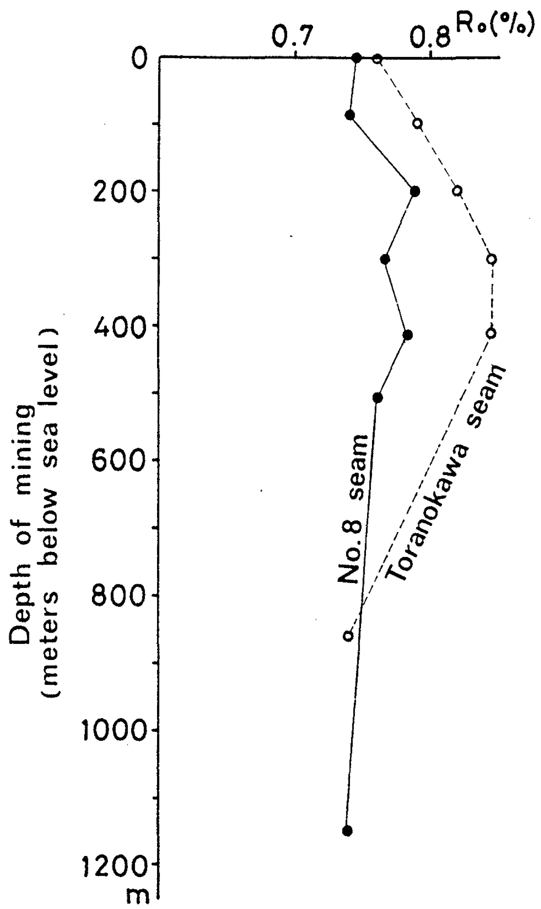


Fig. 11. Changes in the rank of coal of No. 8 and Toranokawa seam with depth of mining in the Mitsui Ashibetsu colliery.

1970; BOSTICK, 1974; HOOD, GUTJAHR and HEACOCK, 1975). BOSTICK's hydrothermal bomb experiment shows that vitrinite reflectance was not changed in different runs where the pressure was varied keeping temperature constant, and support that the pressure does not progress coalification. Based on his experiment he established "Well-standard" line for the estimation of the paleo-temperature from reflectance of samples buried roughly for 30 to 80m.y. (Fig. 12).

Comparison was made on the Japanese Paleogene coal using this line. The Japanese Paleogene coal is Oligocene or Eocene in age and is considered to reach the maximum burial stage sometime in Miocene and, therefore, to bury for 30 to 45 m.y. The samples for the comparison are taken from Table 1 in which the relation between reflectance and fixed carbon content of the sample is reasonably reconcile with the aforementioned correlation curve between the reflectance and the fixed carbon content (Fig. 2).

The temperature on the samples measured from the "Well-standard" line (Fig. 12) is listed on Table 4. As mentioned in the previous chapter, the critical reflectance to separate the coking bituminous coal from the non-coking subbituminous coal is about 0.6. Temperature equivalent to 0.6 of reflectance is about 80°C. From the table it can be estimated that the formation temperature of the coking bituminous coal ranges from 80° to 115°C and that of the non-coking lignite or subbituminous coal is as much as 80°C, may be less 60° to 75°C.

Paleogeothermal Gradient in the Paleogene Coalfields

In previous chapters the temperature of coal formation are estimated by two different ways, one by zeolitic reaction temperatures and the other from BOSTICK's. They are closely agree with each other for each sample. They are approximately accepted as the temperature at the depth of maximum burial which took place sometime in Miocene in the Japanese Paleogene coal-

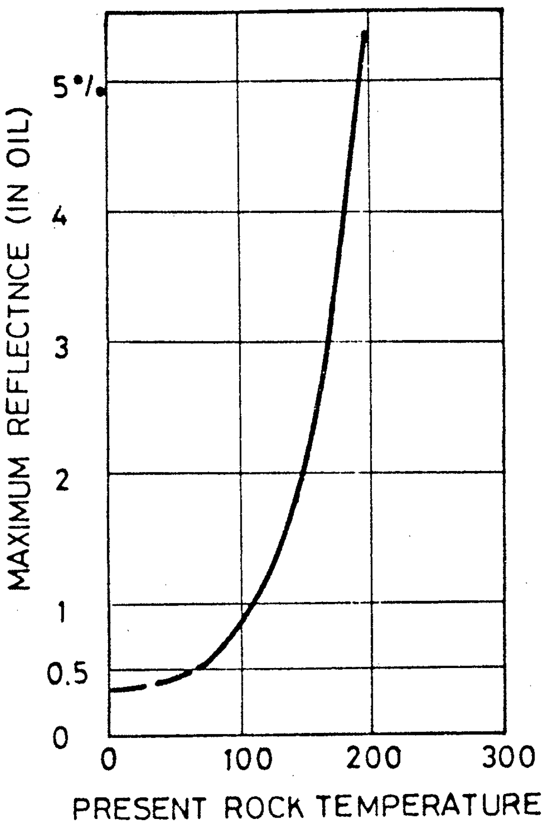


Fig. 12. BOSTICK's "Well-standard" line.

Table 4. Measured temperature from the BOSTICK's "Well-standard" line for the Japanese Tertiary coal.

Locality	Formation	Coal rank	Temperature
Tempoku	Soya	lignite	60
Joban	Iwaki	"	70
Kushiro	Harutori	Subbituminous	75
Takamatsu	Onga	bituminous	88
Bibai	Bibai		90
Akabira	"		92
Ashibetsu	"		94
Miike	Toka		94
Yubari	Yubari		100
Takas hima	Hashima		110

fields.

If the depth of maximum burial is estimated as 5,000 m in the Ishikari coalfield, 3,000 m in the Kushiro and 2,000 m in the Joban and Miike, and the surface temperature is assumed at 13°C in Hokkaido, and 15°C in others, the calculated paleogeothermal gradients are 1.6°C per 100 meters for the Ishikari coalfields 2.5°C for the Kushiro, 2.7°C for the Joban, and 4°C for the Miike.

The calculated paleogeothermal gradient is little lower in the Ishikari coalfield than the present geothermal gradient but in general is similar to the present one.

Summary

1. The Japanese Tertiary coal ranges from lignite, through subbituminous coal to bituminous coal. (Anthracite is only locally limited to the aureole of igneous intrusives so that not discussed in this paper).

2. Authigenic zeolites occur exclusively as an alteration product of silicic glass in vitric tuffs which are sparsely interbedded with coal measures. These authigenic zeolites found in the Japanese coalfield are distributing systematically in a vertical zonal arrangement within a limited local stratigraphic succession. Such zeolite zoning is essentially the same as the zoning in the oilfield. In the Tertiary oilfield, the study on the most typical present-day burial zeolitic diagenesis cleary shows that the alkali zeolite reaction is essentially temperature dependent.

3. The rank of coal is specifically correlated with zeolite zone throughout the Japanese coalfields. All of the lignite and subbituminous coal occur within Zone II (clinoptilolite-mordenite) whereas the bituminous coal occurs exclusively with Zone III (analcime).

4. It is, therefore, reasonably concluded that the coalification of the Japanese Tertiary coal is essentially temperature dependent and it is estimated that the formation temperature of the lignite and subbituminous coal are as much as about 80°C while that of the bituminous coal ranges approximately from 80°C to 125°C.

5. The studies in the Mitsui Sunagawa and Mitsui Ashibetsu areas show that irregularities in the rank of coal with stratigraphic position are recognized, even in the limited area. Such irregularities are explained that the degree of coalification was sensitively affected by the lithology of the host rock, in other words by the irregularities of temperature gradient.

6. They also show that the rank of coal does not change with an increase of the depth of mining (0 m ~ −1.000 m). It is likely that the post deformational depth of burial gave little effect on the rank of coal so that the retrogressive coalification is not apparent.

7. It is much emphasized that the temperature is main factor to control coalification, which was provided at the depth when buried since duration of burial is geologically interpreted that it was brief in the coalfield studied.

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古第三紀石炭の石炭化作用に及ぼす温度の影響

下　山　俊　夫・飯　島・　　東

（要　　旨）

筆者等は先に本邦古第三紀の主要炭田における炭質，埋没深度，構造，沸石の産状を検討し，石炭化作用を規制する主要因は，埋没続成時の地温勾配であると推論した (SHIMOYAMA and IIJIMA, 1976；下山・飯島, 1977). すなわち大局的に見れば炭質と続成沸石帯はよく対応し，褐炭，亜瀝青炭は，II 帯に，瀝青炭は，III帯に属することより推論される．

石炭化作用は熱履歴つまり温度と時間に依存すると一般に言われているが，隆起時或いは隆起後 (retrogressive) の石炭化度の進行は実際上は認められない．三井芦別においては0～－1,000m の採掘深度の範囲で，虎皮層等特定の炭層の石炭化度は殆んど変化しない事から推論される．

一方，局地的に見れば石炭化度は夾炭層の岩相に影響される．例えば三井砂川，三井芦別において，下位の炭層が必ずしも上位の炭層より石炭化度が高くない事は，前者が比較的厚い砂岩帯に夾有されていることで解釈される．