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Genesis of the Paleogene Purple-Red Beds in Western Kyushu, Japan*

Takashi MIKI and Hiroharu MATSUEDA**

Abstract

Purple-red beds located at the basal parts of the Paleogene sedimentary sequences in western Kyushu, Japan, were lithologically and mineralogically investigated. The results indicate that these beds were deposited in small depressions which transformed gradually to coal swamps. The pigmenting agent responsible for this characteristic color is hematite that fills interstices among the grains. The hematite was supplied from schists and igneous rocks as detrital fine mineral grains and from ground water as hydroxide that aged to hematite through diagenetic dehydration. The formation of the overlying coaly sediments may also have genetical relations to the origin of color.

I. Introduction

Red beds in many districts of the world have attracted the attention of many researchers not only because of their characteristic color but also their sedimentological significance (VAN HOUTEN, 1948, 1968; JONES, 1965; TOMPSON, 1970; IIJIMA, 1972; and others). Some investigators suggested that the sediments might be used as a paleo-climatic indicator (VAN HOUTEN, 1961; POWER, 1969) and others studied the mineral transformation of iron oxides during diagenesis (SCHMALZ, 1959; BERNER, 1969). Syn- and epigenetic theories have been proposed for the origin of red beds (TOMPSON, 1970; SLÁNSKÁ, 1976).

In western Kyushu, Japan, purple-red beds*** occur in the basal parts of the coal-bearing Paleogene sequences. Due to their color, the presence of the sediments provides striking contrasts to the under- and overlying formations. However, no detailed studies have been done on these sediments. We have been investigated these characteristic sediments paying special attention to a relationship between the sediments and accompanying coal-seams. In this paper we attempt to provide a consistent theory on the genesis of the colored beds in western Kyushu after characterization of the sediments in each basin based on the previously reported data in Amakusa (MIKI and MATSUEDA, 1974) and Takashima (MIKI and MATSUEDA, 1978) and newly obtained ones in Ōmuta and

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*** Although a term "purple-red" does not express exactly the color of these sediments, the term is customarily used in this paper following the previous works (NAGAO, 1926; MATSUSHITA, 1949).

Sakito. The preliminary descriptions are presented also from a paleomagnetic viewpoint.

This is a part of the sedimentological studies of coal-bearing sequences in Kyushu, and a contribution to the IGCP Project No. 166, "Correlation of Coal-bearing Formations". This paper is dedicated to Prof. Tsugio SHUTO in the occasion of his retirement from the professorship at Kyushu University.
Acknowledgements: We would like to acknowledge the encouragement of Prof. Kazuo NORITOMI of Akita University during the course of geomagnetic study. Mr. Jun-ichi WATANABE performed geomagnetic measurement of rocks. We express our thanks to Prof. Kiyoshi ISHIBASHI of Fukuoka University and to Mr. Tsugio HIRANO of Akita University who kindly analyzed the rock samples. We are deeply indebted to Dr. Hideomi KODAMA of Chemistry and Biology Research Institute, Agriculture Canada, for critical reading of the manuscript. The present study was financially supported in part by a Grant-in-Aid from the Ministry of Education, Japan.

II. Areal and stratigraphical distribution of the purple-red beds

Coal-bearing Tertiary sediments are distributed in more than ten different places in Kyushu. In western Kyushu the Paleogene formations that accompany

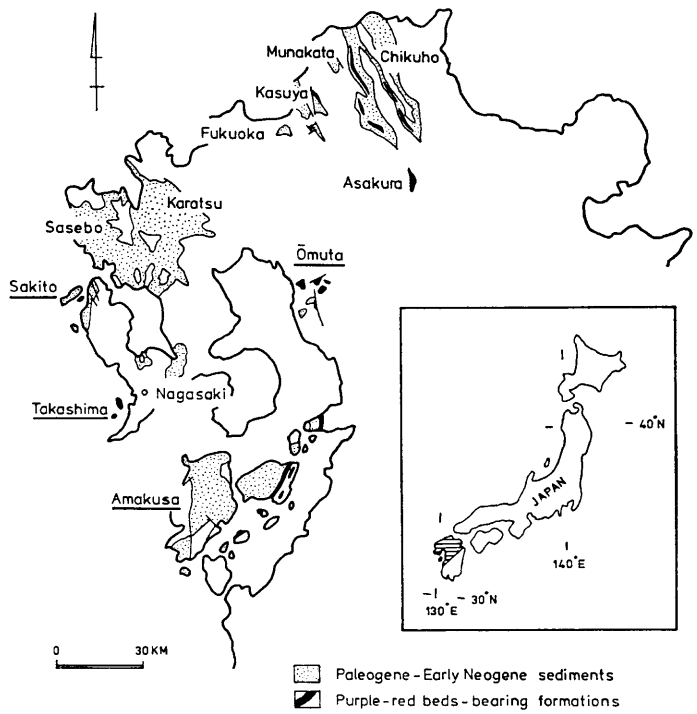


Fig. 1. Distribution of the Tertiary coal-fields and the purple-red beds-bearing formations in Kyushu. Areas studied in this paper are underlined.

the purple-red beds are found in the four small basins of Ōmuta, Amakusa, Takashima and Sakito from east to west (Fig. 1). Paleogene sequences in each basin are divided into some groups and formations and correlated with one another(Fig. 2).

The lowest member that contains age-determinable fossils is the Shiratake Formation in Amakusa and its equivalent in Ōmuta from which the early Eocene foraminifers and molluscs are found, and the Kōyaki Formation in Takashima

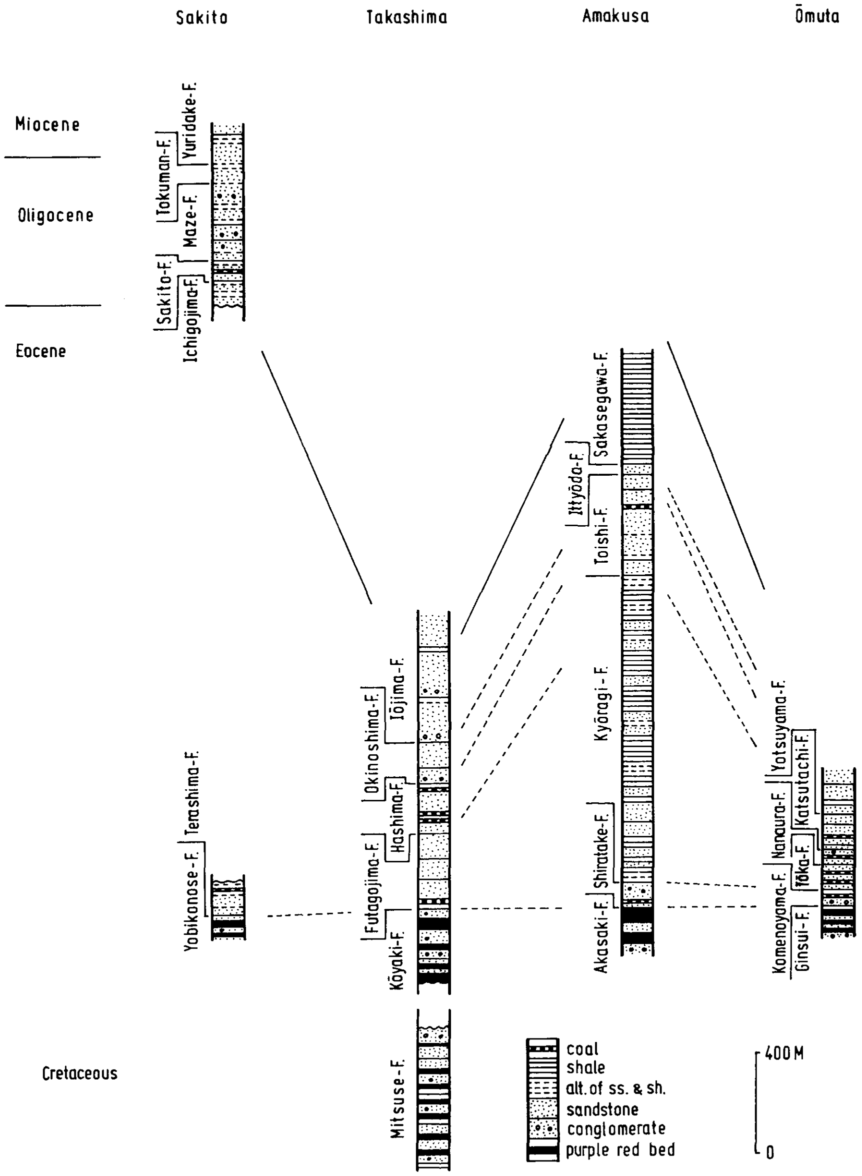


Fig. 2. Generalized columnar sections of the Tertiary formations and their correlations. (after MIKI, 1975; MIKI and MATSUEDA, 1978).

where some molluscs occur. The age of the uppermost part of the Tertiary sequences in the Sakito district ranges to Miocene.

In most places the workable coal seams are intercalated in the middle parts of the sequences. Unworkable coaly sediments occur just above the purple-red beds-bearing formations. Purple-red beds are selectively contained in the basal parts of the Tertiary named the Akasaki Group. The upper Cretaceous sequences in Takashima, which underlie the Paleogene sediments without the remarkable lithological change, contain exceptionally plenty horizons of purple-red beds. On the other hand, in the Yobikonose Formation of the Sakito district, a lesser amount of such colored rocks is contained in comparison with other three basins.

III. Field and laboratory data

A. Mode of occurrence

The formations of purple-red beds consist of conglomerate, sandstone and shale, and among them, shale and/or mudstone selectively show characteristic purple-red color (very dark red, 5R 2/6; blackish red, 5R 2/2; and grayish red, 5R 4/2; according to the rock-color chart of the Geological Society of America). Sand-sized purple-red sediments are rarely found. Clayey matrices of the basal conglomerates directly overlying the basement rocks sometimes show the purple-red color. The thickness of a purple-red layer changes remarkably from place to place: from 1.5 m to 2.5 m in Ōmuta, and from 2 m to 10 m in Takashima for example. The changes of thinkness and lithology over a short distance are also observed in the exposures (Fig. 3B). The purple colored parts often contain the irregularly shaped drab mottlings which generally vary from one to several centimeters in diameter.

Modes of occurrence of the characteristic beds are macroscopically classified into four types as already discussed (MIKI and MATSUEDA, 1974, 1978).

Type A: The purple-red and drab beds are alternately superimposed with distinct contact planes. The color boundaries are roughly parallel to bedding planes (Fig. 3B).

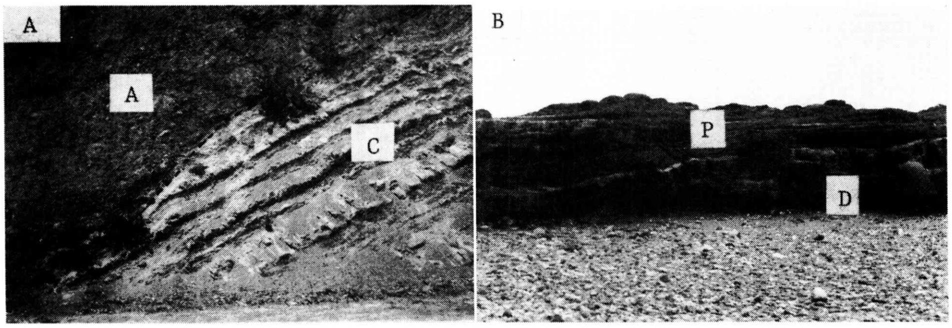


Fig. 3. Exposures of the purple-red beds. (A) Unconformity plane (broken line) between the Cretaceous (C) and the Paleogene purple-red beds-bearing Akasaki Formation (A) observed in Mae-jima, Amakusa district. (B) Alternation of the purple-red beds (P) and drab ones (D) in Uto, Amakusa district (Type A).

Type B: The purple-red parts accompany the drab mottlings showing a fragmental and pebble-like appearances. The boundaries between both parts are clearly recognized.

Type C: The drab parts diffuse into the purple areas. The distinct contact planes can not be observed. The mode of occurrence of this type is most frequently found.

Type D: The purple-red areas are partly bleached to grayish blue color along the cracks.

Type A and C may correspond to that of 1 and 2 of TURNER (1980)'s classification respectively. He also recognized the additional occurrence similar to the Type D. As stated before, the beds in Amakusa and Takashima show chiefly the characteristics of Type A and C respectively. The purple-red beds in Ōmuta and Sakito show mainly, on the other hand, those of Type C and D, and Type A respectively.

Pebbles of conglomerates are poorly sorted and angular in shape.

B. Petrography and mineralogy

Conglomerates contained in the purple-red beds-bearing formations consist of pebbles and cobbles of mica and hematite-bearing crystalline schists, chert, granitic and volcanic rocks as well as sedimentary rocks. Petrographical characters of sediments in Amakusa and Takashima are summarized as follows after previous works (MIKI and MATSUEDA, 1974, 1978). In the Akasaki Formation of Amakusa, metamorphic pebbles predominate, whereas in the Kōyaki Formation of Takashima, igneous pebbles are abundant. The igneous pebbles contain detrital grains of iron minerals showing ilmenite-magnetite exsolution texture which is indicative of igneous rock origin. Heavy and light-mineral compositions of sandstones in two basins (MIKI, 1975) support the observations

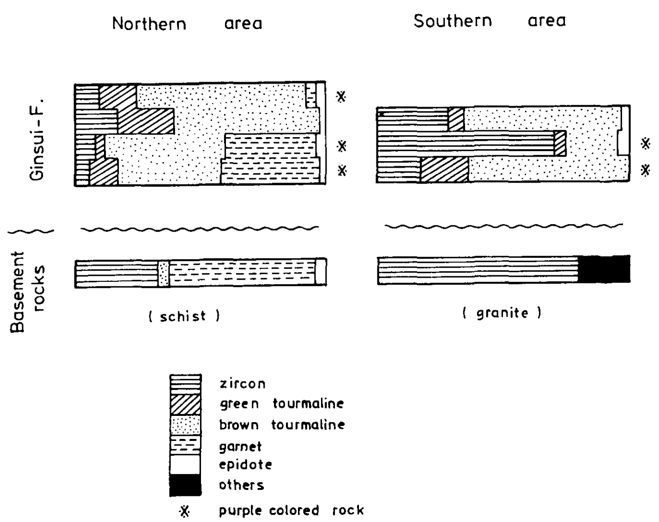


Fig. 4. Heavy-mineral assemblages of the Tertiary and its basement rocks in Ōmuta.

mentioned above. Epidote, a characteristic heavy-mineral species in pebbles of schist in conglomerates, is found abundantly in sandstones.

The recent investigations in Ōmuta and Sakito indicate an interesting geological phenomenon. In the Ginsui Formation of Ōmuta, the beds overlying granitic rocks in the southern area show heavy and light-mineral assemblages similar with those of basement granite. Heavy-mineral associations of the beds accompanying mica-rich sandstones, which overlie metamorphic rocks in the northern district, resemble to those of metamorphics (Fig. 4). The Yobikonose Formation of Sakito which rests on granitic rocks contains predominantly the granitic pebbles besides the pebbles of chert.

The characters of sediments described indicate that the source materials, which are clearly different from those of the over- and underlying drab formations, were supplied from the adjacent land areas to the purple-red beds-bearing formations in each basin with short distant transportation.

Although the different mineral compositions can be recognized between the purple and drab parts of the sediments of Type B, no differences are found in the samples of Type A and C. A sole difference between the both colored parts in the purple-drab pair observable under the polarizing microscope is that the purple pigments filling up the interstices of the matrices which occur only in the purple area. The pigments are identified as fine grained hematite by X-ray, reflecting microscope and EPMA analyses. Detrital hematites in different sizes are found in the both colored parts (Fig. 5B). Fine grained hematites are also produced along the cleavages of clay minerals in some samples. A small amount of magnetite are partly martitized, and so-called limonite are also found in the both colored areas.

In general, mineral compositions in the colored parts from various localities are similar as indicated by X-ray diffraction data. Illite, chlorite, illite-montmorillonite mixed-layer mineral and kaolinite were detected as clay minerals.

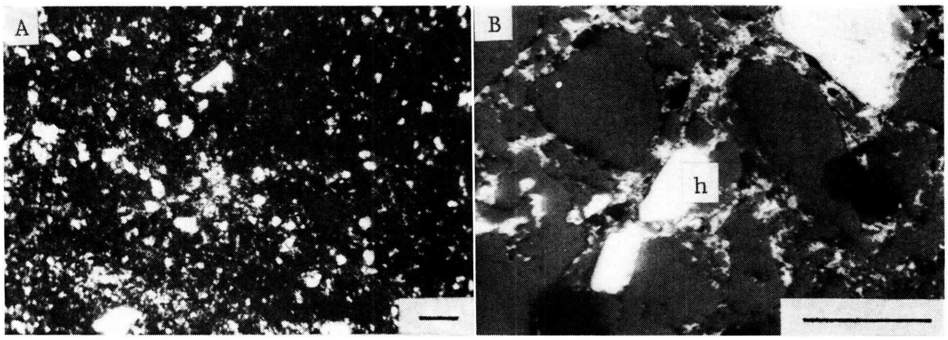


Fig. 5. Photomicrographs of samples. (A) Photomicrograph under polarizing light. Purple-red part (right half) and drab part (left half). (Type C). Crossed nicol. Scale bar is 0.1 mm. (B) Photomicrograph under reflecting light. Hematite occurs as detrital grain (h) and in matrices. Scale bar is 0.1 mm.

C. Geochemistry

Similar to the geochemical data reported from various areas in the world

Table 1. Partial chemical analysis of the purple and drab sedimentary rocks from Ōmuta. Concerning the analytical data for specimens from Amakusa and Takashima, see MIKI and MATSUEDA (1974, 1978).

	1p	1d	2p	2d	3p	3d	4p	4d	5p	5d
TiO_2 (wt. %)	0.49	0.61	0.57	0.60	0.51	0.63	0.57	0.68	0.57	0.63
Fe_2O_3	5.46	2.16	6.98	1.80	5.77	1.94	6.22	2.37	4.62	3.22
FeO	0.68	0.67	0.74	0.54	0.54	0.81	0.39	0.47	0.82	0.59
MnO	0.06	0.06	0.04	0.02	0.03	0.03	0.02	0.01	0.05	0.06
Na_2O	2.10	1.96	1.10	1.31	1.18	1.37	0.30	0.22	0.91	0.84
K_2O	2.49	1.99	3.01	2.64	2.89	2.29	2.41	1.94	2.89	2.79
Fe_2O_3+FeO(wt.%)	6.14	2.83	7.72	2.34	6.31	2.75	6.61	2.84	5.44	3.81
Fe_2O_3/FeO	8.03	3.22	9.43	3.33	10.69	2.40	15.95	5.04	5.63	5.46
Na_2O/K_2O	0.84	0.98	0.37	0.50	0.41	0.60	0.12	0.11	0.31	0.30

p: purple-red, d: drab. Analyst: K. ISHIBASHI

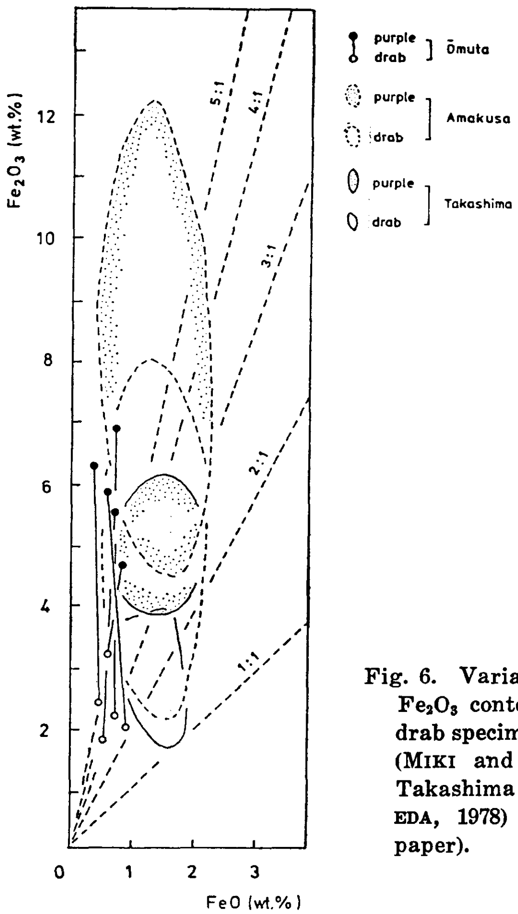


Fig. 6. Variation of FeO and Fe_2O_3 contents in purple and drab specimens from Amakusa (MIKI and MATSUEDA, 1974), Takashima (MIKI and MATSUEDA, 1978) and Ōmuta (this paper).

(VAN HOUTEN, 1961; LOUGHNAN, et al., 1963; SLÁNSKÁ, 1976; and others), wet chemical analysis of the present samples shows high contents of ferric and ferrous irons, particularly in the Amakusa district as shown in Fig. 6*. Ferric iron contents of materials in Ōmuta are not so different from those in Takashima. Judging from the results of petrographic and X-ray examinations, it is considered that FeO is chiefly due to chlorite, magnetite and biotite, and Fe_2O_3 to hematite, limonite, magnetite and epidote.

Unlike the FeO contents, Fe_2O_3 content varies widely and its maximum content reaches to 12.21% in the purple-red part. Fe_2O_3/FeO ratios of the purple parts are higher than those of the drab parts in paired specimens. Concentration of Al_2O_3 and Fe_2O_3, and leaching of SiO_2, K_2O and Na_2O as observed in many lateritic red beds were not recognized in the present materials.

D. Rock magnetism**

Paleomagnetic study was carried out on specimens from Amakusa, Taka-

Table 2. Magnetic susceptibility (emu/g·Oe) and intensity of remanent magnetization (emu/g) of purple-red specimens from Amakusa (Shiratake and Akasaki Formations, and Himenoura Group) and Ōmuta (Ginsui Formation).

Magnetic susceptibility

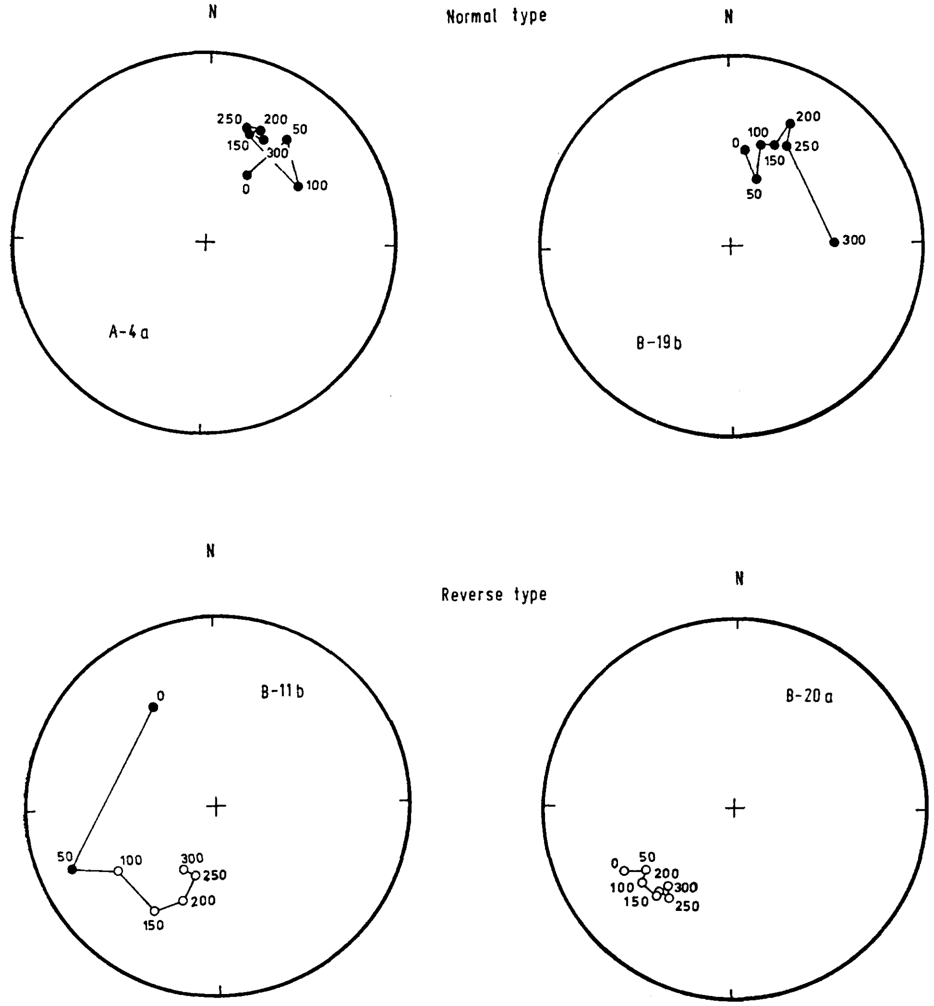
Age	Stratigraphy	No. of samples (N)	Susceptibility (log κ)	Std. dev. (σ)
Tertiary	Shiratake F.	6	−6.53	0.189
	Akasaki F.	24	−5.90	0.398
	Ginsui F.	5	−6.29	0.019
Cretaceous	Himenoura G.			
	(Upper)	6	−6.15	0.198
	(Middle)	3	−6.17	0.180
	(Lower)	1	−6.37	—

Intensity of remanent magnetization

Age	Stratigraphy	No. of samples (N)	Intensity (log J)	Std. dev. (σ)
Tertiary	Shiratake F.	17	−6.37	0.193
	Akasaki F.	18	−5.43	0.315
Cretaceous	Himenoura G.			
	(Upper)	17	−6.00	0.514
	(Middle)	8	−5.73	0.660
	(Lower)	1	−6.04	—

* Relationships between the chemical compositions of samples and their modes of occurrence were described in the previous papers (MIKI and MATSUEDA, 1974, 1978).

** Experimental procedures and the detailed results of the investigations will be described in another paper.



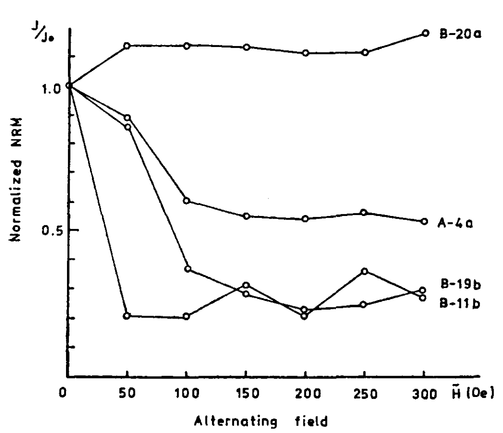


Fig. 7. A.C.-demagnetization and their pole projection on SCHMIDT's net of specimens from the Akasaki Formation in Amakusa. Solid and open circles indicate normal and reversed magnetizations respectively.

shima and Ōmuta in order to examine the magnetic properties of colored rocks and their availability for the geological consideration.

Magnetic susceptibilities and intensities of remanent magnetization of materials from Amakusa and Ōmuta are shown in Table 2. As is evident from the table, purple-red sediments are more strongly magnetized than the drab samples of the over- and underlying formations. High intensities of remanent magnetization after alternating field demagnetization in 50 Oe peak field are also recognized for the purple-red specimens.

Curie temperature obtained from thermo-magnetic curve, most of which are 540°C to 560°C, indicate that the most important remanence carrier is not

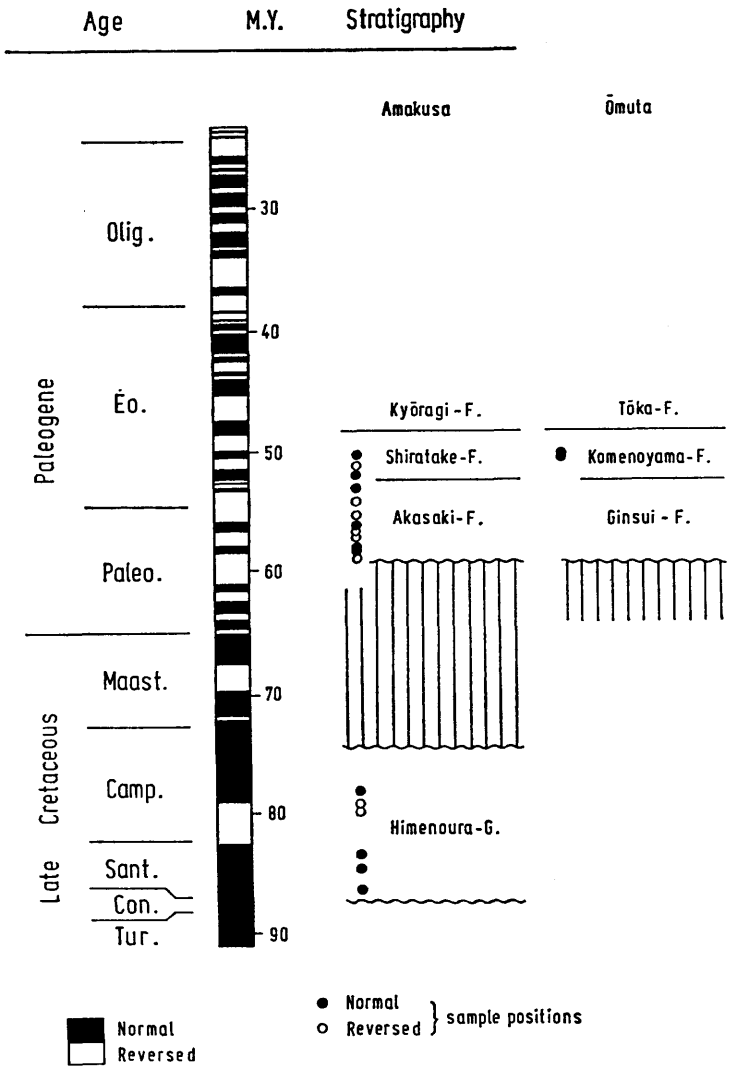


Fig. 8. Magnetostratigraphy in the Amakusa and Ōmuta districts. Standard time scale is after HARLAND et al. (1982).

pigmentary hematite but magnetite or titanomagnetite. Hematite which is observed under the microscope does not contribute for rock magnetism probably due to its small grain size and/or its poor crystallinity. This experimental result is in harmony with the microscopic observation showing the coexistence of fine grained hematite and coarse grained detrital magnetite.

OZIMA and OZIMA (1972) claims that natural remanent magnetization of red rocks is originated from remanent magnetization of hematite secondarily attached by chemical alteration. However, strong magnetization of the present materials in western Kyushu does not due to hematite but to detrital magnetite which involves depositional remanent magnetization as already mentioned. Consequently, paleomagnetic discussions could be available on the data obtained in this study.

Samples from the Akasaki Formation in Amakusa show the magnetizations of normal (NE declination) and reversed (SW declination) polarities (Fig. 7). This fact does not always support an idea of the so-called Akasaki Normal Epoch proposed by SASAJIMA et al. (1968). Preliminary work on the magnetostratigraphy using colored and associated drab sediments suggests that the purple-red beds-bearing formations in Amakusa including no fossils* range from middle Paleocene to early Eocene (Fig. 8).

Special attentions should be paid to the fact that the geological ages of the purple-red beds sporadically distributed throughout Kyushu are different in each basin: Cretaceous in Takashima and Oligocene in northern Kyushu, for example. Paleomagnetic study on the colored materials will provide the useful data for the future investigations of the characteristic beds in these basins.

IV. Sedimentary conditions

Judging from an appearance of the poorly-sorted and angularly-shaped basal conglomerate, remarkable lateral change of thickness and litho-facies of the formations, high quantities of heavy minerals in sediments, and those of hematite in spite of absence of siderite and pyrite, the colored beds-bearing formations were considered to be formed in a small, shallow, and tectonically unstable depositional basins. Shallow sea environment was proposed by JONES (1965) for the Cretaceous red beds in Nigeria. BERNER (1971) illustrated the depositional environment of the red beds as river flood plain adjacent to land. It is well known that a remarkable crustal movement took place prior to the sedimentation of the Paleogene formations in western Kyushu (MIKI, 1975), and this movement acted as an initiator to make the depressions as a result of a local differential subsidence of the lands in which the formations are accumulated. It is easily understood that the formations show the earliest stage in the cycle of sedimentation, and that the topographical relief beneath and around the basins were still preserved.

Geochemical and petrographical studies revealed that the source materials

* The age of the basal part of the overlying Shiratake Formation is regarded as Ypresian from fossil evidence (MIKI, 1975).

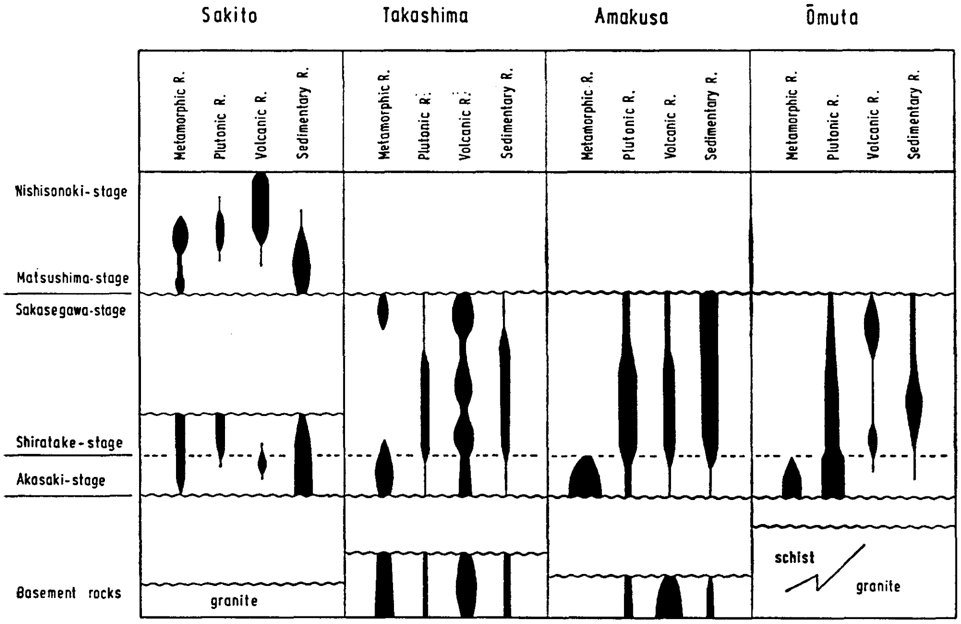


Fig. 9. Idealized figure illustrating the transition of clastics supplied to the basins.

forming the purple-red beds-bearing formations were derived chiefly from metamorphic and igneous rocks exposed near the depositional sites not only in Amakusa and Takashima as previously reported (MIKI and MATSUEDA, 1974, 1978) but also in Ōmuta and Sakito as described in this paper. A marked transition of the clastics for the overlying drab-colored formations into the predominant amounts of sedimentary rocks of older ages instead of metamorphics (Fig. 9) suggests that the metamorphic materials transported to the depositional sites played an essential role for coloring of the sediments.

Above petrographical and geochemical descriptions support the paleogeographical environment previously illustrated (MIKI, 1975, Fig. 36).

V. Discussion on the genesis

Several hypotheses have been proposed concerning the origin of the red color of some ancient beds for more than 100 years. As summarized by TURNER (1980), early workers thought that the red beds were formed under desert conditions. KRYNINE (1949) considered that the red beds were derived directly from the red colored lateritic soils. WALKER (1967) attached importance to a diagenetic processes for coloring of the beds.

The purple-red beds in the present areas show no features commonly observed in seatearths or paleosols such as root scars. Modes of occurrence and chemical compositions of these beds do not give any evidence of their lateritic origin in situ. The iron substances were supplied from basement complex in various forms such as iron hydroxides (so-called limonite), iron-bearing silicate minerals, etc, and ultimately transformed into hematite that is responsible for coloration. The result of geomagnetic study indicates the transportation also of magnetite to the depositional sites. The existence of pebbles and fragments of hematite-bearing schist in the purple-red sediments indicates that a part of the pigments might have been derived from finely divided hematite crystals. The beds in Amakusa were derived from a great amount of metamorphics. They contain higher irons compared with those in other districts. The purple-red beds are inserted in the formations consisting of clastics originated from iron-rich metamorphic and igneous rocks. These facts support the above mentioned presumption.

The former work (MIKI and MATSUEDA, 1978) showed an existence of a relation between the amount of purple matrix and grain size of specimens on samples from Takashima. This relation was obviously recognized also on samples from other three basins in this paper (Fig. 10). This figure indicates that the finely divided hematite and limonite were primarily supplied and deposited with accompanying detrital clay fractions. When such iron-rich materials are accumulated alternately with ordinary iron-poor clastics, purple-drab paired layers (Type A) whose color boundary roughly corresponds to bedding planes are accomplished.

Concerning the genesis of the drab mottlings scattered in the purple-red sediments (Type C), many workers currently insist that the latter changed to

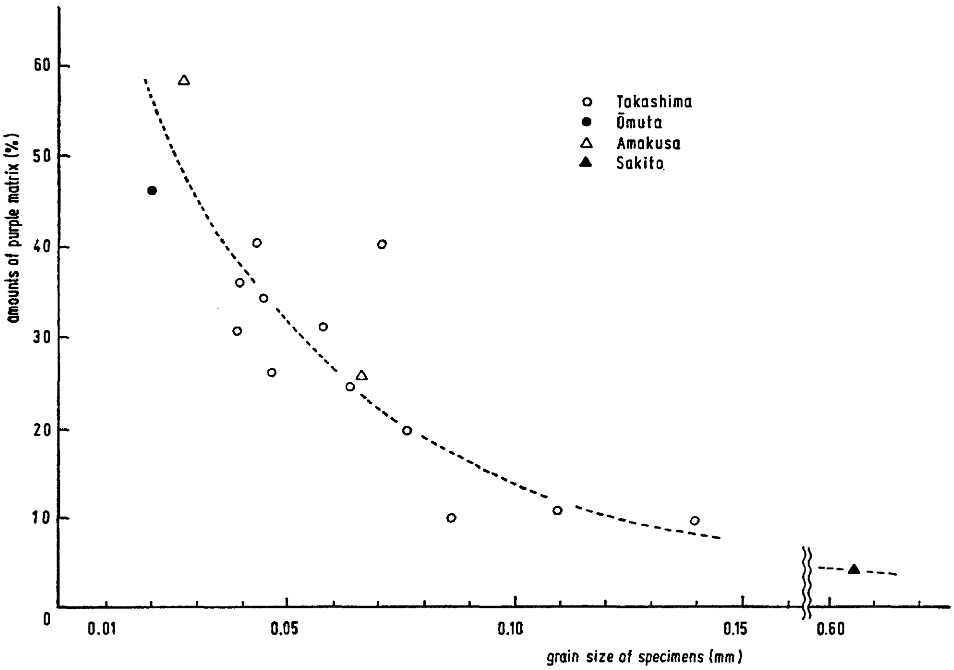


Fig. 10. Relationship between the amount of purple matrix and grain size of rocks. Data of Takashima are after MIKI and MATSUEDA (1978).

the former by partial reduction after sedimentation due to existence of organic matter. TURNER (1980) described that the reduction spots contained more calcium carbonate and less clay matrix than their host red sediments. We find, however, no positive indications to support this proposal in the present areas such as organic remain, high contents of $CaCO_3$ and lesser amount of clay matrix in the drab parts compared with the surrounding purple-red sediments. The total iron contents of the drab spots are less than those in the host purple-red parts in spite of the nearly similar level of the FeO between the two. This is not compatible with partial reduction theory of the drab mottlings, because of reduction iron becomes soluble. Our present explanation for the origin of the color is based on the idea that differential attachments of irons from ground water by heterogeneous soaking to the sediments.

In the overlying formations with no purple-red beds of four basins some

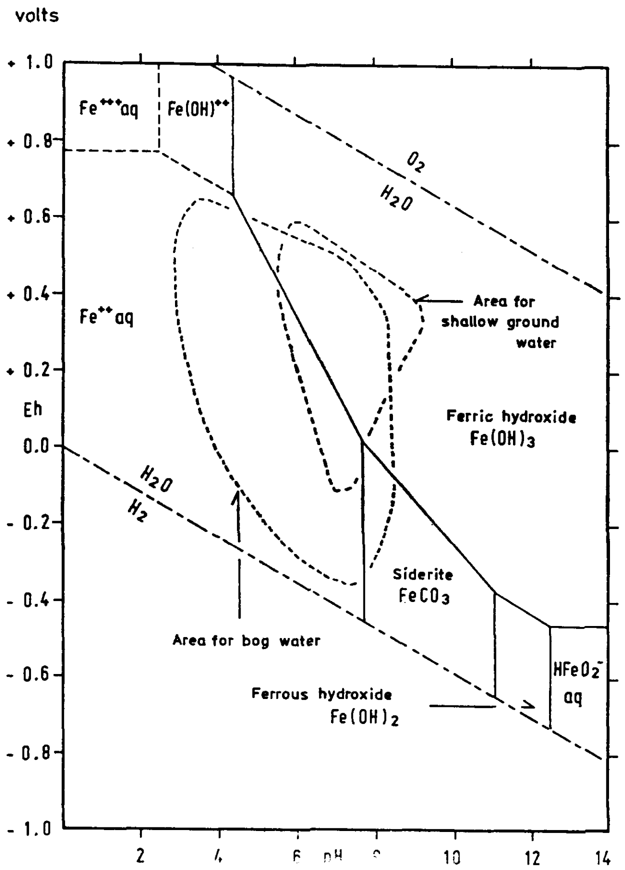


Fig. 11. Stability field diagram for iron substances at 25°C and 1 atmosphere total pressure. Boundary between solids and ions at total activity of dissolved species $=10^{-6}$. Total dissolved carbonate species $=10^{-2}$. (after BAAS BECKING et al., 1960; GARRELS and CHRIST, 1965; WALKER, 1967).

coal seams or coaly shales are intercalated without an exception (Fig. 2). This easily leads a presumption that the ground water of low pH and low Eh percolated among the sediments during the sedimentation of coaly materials and, in other words, during the diagenetic alteration of the underlying purple-red beds, as is known in the present-day coal swamps. Such ground water dissolved a plentiful ferrous iron primarily supplied and transported it to the depositional sites in various forms. When the pH and/or Eh of ground water ascend in a later stage by marine water invasion, uplifting of the basin floor, and other reasons, the dissolved ferrous iron may precipitate as an iron hydroxide, namely limonite, which was transformed to hematite by dehydration under diagenetic conditions. In this viewpoint, accumulation of coaly sediments is regarded to have played an important role for the coloration of the beds.

Judging from the occurrence of martite and fine-grained hematite along the cleavages of clay minerals, diagenetic alterations of iron oxide and iron-bearing silicates also contributed for coloration of the sediments. It is probable that a part of iron was attached to or dissolved from the sediments by ground water in other later stage.

In summary, our hypothesis proposed for the origin of purple-color may be schematically expressed as shown in Fig. 12. This suggests that the purple-red color observed today reflects the duplication of some causes occurred syn- and epigenetically.

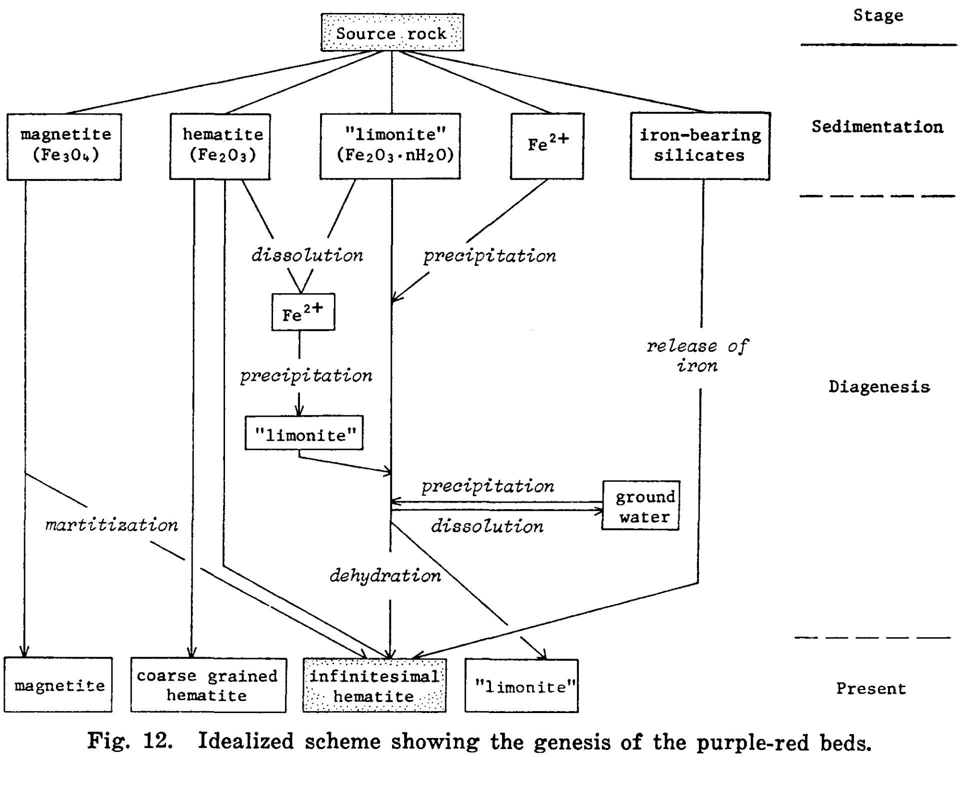


Fig. 12. Idealized scheme showing the genesis of the purple-red beds.

The present study reveals that the purple-red beds in four basins show the characteristic genesis and depositional environment in the earliest stage of the Paleogene sedimentation. Similar colored beds are distributed also in other basins throughout Kyushu always accompanying coal seams (Fig. 1), and their genetical discussions are left untouched due to the insufficient data. They will be continuously studied from a special viewpoint of the genetical relation between the purple-red beds and coal seams as described in this paper.

VI. Concluding remarks

Two conditions are necessary for purple-red coloration of these beds. The first is a supply of a great amount of iron in various forms. This means a preservation of a topographical relief consisting of iron-rich basement rocks near the depositional sites in the earliest stage of a cycle of sedimentation. The second is a movement of iron in sediments under sedimentary and diagenetic conditions, namely dissolution of iron into ground water and attachment of iron into sediments. Low pH and Eh of ground water during the accumulation of coaly sediments are responsible for the conditions required.

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