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**REPORT No. 230**

GEOLOGICAL SURVEY OF JAPAN

# GENESIS OF THE FIRECLAY DEPOSITS IN TAJIMI-TOKI DISTRICT, GIFU PREFECTURE, CENTRAL JAPAN

By

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GEOLOGICAL SURVEY OF JAPAN

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Konosuke SATO, Director

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Abstract

A few kinds of fireclay deposits, which are kibushi-clay, gaerome-clay and the so-called kaolin, are developed in the earlier Pliocene Tokiguchi formation of this district. This formation appears to have been deposited separately in several basins. And its sedimentary features in each basin are not always similar, and appear to reflect closely the basement geology. The distribution of the fireclay deposits would have been remarkably controlled by the differences of the basement geology and sedimentary process of the Tokiguchi formation in each basin.

Each of these fireclays has its own characteristics concerning fabric, mineral composition, type of main kaolin mineral, refractoriness and so on. Kibushi-clay generally occurs accompanied by lignite seams. It is composed mainly of disordered kaolinite and fine-grained quartz with montmorillonite and a small amount of illite. Gaerome-clay, which presumably was derived from weathered granite, consists largely of kaolin minerals and quartz grains accompanied by feldspar and mica clay mineral, but its clay fraction percentage is very variable. And the gaerome-clays with large clay fraction contain more kaolinite, and ones with less clay fraction tend to include more halloysite. Such variation in mineral composition of gaerome-clays may have been caused by sorting owing to the difference in particle size of kaolinite and halloysite. Next, the so-called kaolin is composed dominantly of mixture of halloysite and metahalloysite with a small amount of quartz and volcanic glass. Its geological occurrence, petrographical feature and mineral composition show that it had been derived authigeneously from acidic tuff. But kibushi-clay may be mostly detrital originating from weathered granite and other basement rocks. Many geological evidences show that the basement rocks were remarkably subjected to weathering alteration before the deposition of the Tokiguchi formation.

## I. Introduction

Both kibushi-clay and gaerome-clay are representative fireclays in Japan. They are of high refractoriness and are very plastic, and thus have been extensively used as raw materials for ceramic wares. Most of them are produced in the region eastward from Nagoya, which includs Seto-shi and Nishikamo-gun of Aichi prefecture, and also Tajimi-shi, Toki-shi and adjacent districts of Gifu prefecture. At present, about 1,000,000 tons of fireclays are exploited in Japan annually, and two thirds are produced in this region.

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Silica-sand, feldspar and other ceramic raw materials are also mined in this clay-producing region. Therefore, many investigations concerning the geological and mineralogical features of these fireclays have been carried out. Especially their origin has been very controversial and is still not settled.

Above all, with respect to genesis of kibushi-clay, several different conclusions as follows have been reported. KIYONO and ISHII (1926), OSHIMA (1949) and SUDO (1953) assumed that kibushi-clays were secondary sediments derived from weathered granite and were affected to various extent by humic acid arising from lignites. KITAZAKI and ARAKI (1952), AKAMINE (1954) and NOZAWA (1955b) insisted that tuffaceous materials had played a great role as the parent materials of kibushi-clays. But TANEMURA (1963) stated that kaolinite in kibushi-clay was detrital in origin and was presumably formed in hydrothermally altered granite before its deposition.

MATSUZAWA and his collaborators (1960) studied the detailed stratigraphy of the Pliocene Seto group in Seto district, and concluded that there were no positive evidences proving that kibushi-clays originated largely from tuffaceous materials.

Since 1961, the writer has investigated the kibushi- and gaerome-clays in Tajimi-Toki district, with special attention to their geological occurrences and mineral composition. As the result of these investigations, it was showed that the lower Pliocene beds with these clays were distributed separately at scores of depositional basins and their sedimentary features differed with each basin. (FUJII: 1967a) And it was reported that the so-called kaolin which presumably originated from tuff occures in this district and its mineral composition distinctly differs from that of kibushi-clay (FUJII: 1964).

In this paper, the writer presents the results of field studies and laboratory works on these fireclays with particular attention to their sedimentary process and mineral components, and he describes the facts that weathering effects are preserved in the basement rocks overlain by the clay-bearing beds. Moreover some interpretations concerning formation process of fireclay are given.


## Acknowledgements

The writer wishes to express his sincere gratitude to professor Isao MATSUZAWA, Nagoya University, for his kind guidance and advice, and to Professor Hideo TAKESHI, Osaka University, and Dr. Toshio ANSAI, Geological Survey of Japan, for their extensive help and encouragement throughout the entire course of this study. He also thanks profoundly Assistant Professor Keinosuke NAGASAWA, Nagoya University, and Assistant Professor Takeshi UEMURA, Niigata University, for their kind advice.

Furthermore, the writer will never be able to forget valuable assistance of the members of Geological Survey of Japan. Miss Teiko OMORI carried out chemical analysis of many clay samples. Messrs. Akira UNOZAWA and Masafumi INOUE made up the precise topographical-map for this study. Dr. Toshio IGARASHI gave kind assistance for electron microscopic observation. And Dr. Yoshihiko SHIMAZAKI kindly read this paper and reformed construction of the paper.

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Raw-material Co-operation Union, for their kind assistance and guidance during his field work.

## II. Outline of Geology

The clay-producing region is chiefly made up of Palaeozoic system and intrusive granitic and rhyolitic rocks, overlain by Miocene sedimentary rocks, and the Pliocene Seto group is distributed extensively over these bed rocks.

The Palaeozoic system is composed mainly of chert, shale and sandstone disturbed remarkably by intense tectonic movement. Most of the granitic rocks are coarse-grained biotite granite, and the others are granodiorite, diorite etc. The granitic rocks are strongly affected by weathering, and occasionally some of them are made brittle to scores of meters in depth. It is believed that these granitic rocks intruded repeatedly during the period from late Mesozoic to early Palaeogene. (KIYONO and ISHII: 1926, KAWADA: 1961, YAMADA: 1963) And the so-called Nohi rhyolitic rocks are distributed at north of Mizunami.

The Miocene series are distributed in the neighbourhood of Toki and Mizunami, and around Shinano which is the northern part of Seto-shi, overlying these basement rocks unconformably. The Miocene series in Toki-Mizunami district are divided into two groups, the upper Mizunami group and the lower Toki coal-bearing formation. The former consists mainly of marine sediments with abundant tuffaceous material, and the latter of non-marine sediments with a few lignite seams. The Miocene group around Shinano, which is called the Shinano formation, is composed largely of conglomerates and is correlated with the lower Mizunami group (MATSUZAWA and others: 1960).

The Pliocene Seto group is divided into the following two, the lower beds

Table 1 Correlation table of Seto group

<table>
<tr><th>Age</th><th colspan="3">Seto district</th><th colspan="2">Tajimi-Toki district</th><th colspan="2">Aichi and Gifu Province</th></tr>
<tr><td rowspan="3">Pliocene upper</td><td rowspan="4">Seto group</td><td rowspan="3">Yadagawa formation</td><td>Idaka facies</td><td rowspan="4">Seto group</td><td rowspan="3">Tokiguchi formation</td><td rowspan="4">Seto group</td><td rowspan="2">Toki sand and gravel formation</td></tr>
<tr><td>Owari lignite bearing facies</td></tr>
<tr><td>Mizuno sand and gravel facies</td><td>Nagakute sand and mud alternation formation</td></tr>
<tr><td>Pliocene lower</td><td colspan="2">Seto chinaclay formation</td><td>Tokiguchi formation</td><td>Tokiguchi chinaclay formation</td></tr>
<tr><td>Pre-Pliocene</td><td colspan="3">Miocene Shinano formation, Granitic rocks</td><td colspan="2">Miocene Nakamura and Mizunami groups, Granitic rocks, Palaeozoic system</td><td colspan="2">Miocene groups, Granitic rocks, Palaeozoic system</td></tr>
<tr><td></td><td colspan="3">Matsuzawa etc. (1960)</td><td colspan="2">Matsuzawa etc. (1959)</td><td colspan="2">Akamine (1954)</td></tr>
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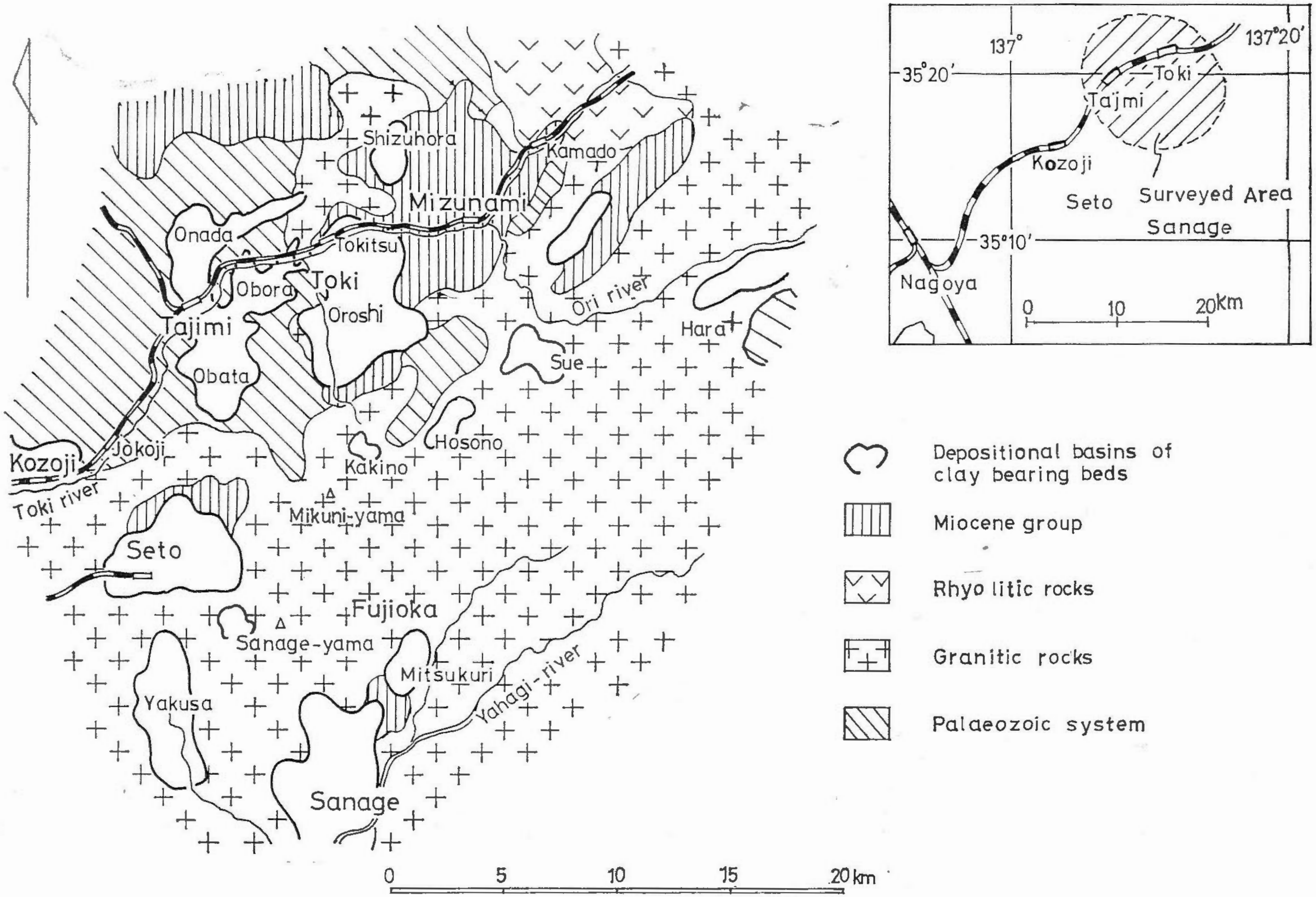



Fig. 1 Paleogeographical map of clay-producing region, Aichi and Gifu Pref., at the earlier Pliocene age

consisting mainly of clay and sand, and the upper ones composed largely of gravel and sand. The lower beds, which are called the Seto china-clay formation or the Tokiguchi formation, presumably were deposited in scores of lakes lying scattered over this region (Fig. 1). Each of these beds, probably of the same age stratigraphically, has different sedimentary features with each other.

The upper beds are named the Yadagawa formation or the Toki formation. They are composed mainly of round gravels of the Palaeozoic and quartz porphyritic rocks cemented by granitic sand. They are non-laminated and developed to 50 meters maximum thickness, and are distributed extensively covering the lower beds disconformably.

It is probable that the upper beds are sediments on flood plane in contrast with the lower ones deposited under calm environment (AKAMINE: 1954, MATSUZAWA etc: 1959, 1960, 1962). Owing to some floral data, the lower beds can be correlated with Pinus Triforia bed (earlier Pliocene), and the upper with the Metasequoia bed (later Pliocene) (MIKI: 1949).

## III. Occurrence of Fireclay Deposits

### III. 1 General features

Most of the fireclay occurring in Tajimi-Toki district are kibushi-clay and gaerome-clay, and moreover the so-called kaolin, which is usually white coloured, is also exploited as fireclay.

The name kibushi-clay means soft and highly plastic underclay with coaly fragments. It occurs accompanied by lignite seams, and is generally blackish brown to dark brown in colour. Good quality kibushi-clay is about SK 34 in refractoriness.

Gaerome-clay is mainly composed of grey coloured clay and fine- to coarse-grained quartz, accompanying a little residual feldspar.

It seems to be poorly sorted sediments derived from weathered granite. Its sorting grade is so variable that it may contain generally 5 to 30 percentages of clay fraction. Gaerome-clay is usually washed by water in order to separate silica sand and clay fractions. These fractions are used as ceramic raw materials, foundry molding sand and others.

The name "kaolin" means white-coloured china-clay being used as raw material for porcelain. It consists mainly of kaolin minerals as well as kibushi- and gaerome-clays. But it is not so plastic as these clays. And it is the notable nature of "kaolin" that it appears white colour when fired.

Good quality "kaolin" contains few impurities and is of high refractoriness ranging from SK 34 to SK 36.

All of the above three kinds of fireclays are intercalated in the Tokiguchi formation, which was deposited separately in Tajimi and Toki basins. The base of Tajimi basin is composed largely of Palaeozoic rocks, and Toki basin consists mainly of granite and Miocene sedimentary rocks.

Tajimi basin may be divided into three sub-basins, which are Onada, Obata and Obora. Connecting with each other by channels, these three sub-basins would have constituted a complicated shape of depositional basin (Fig. 2).

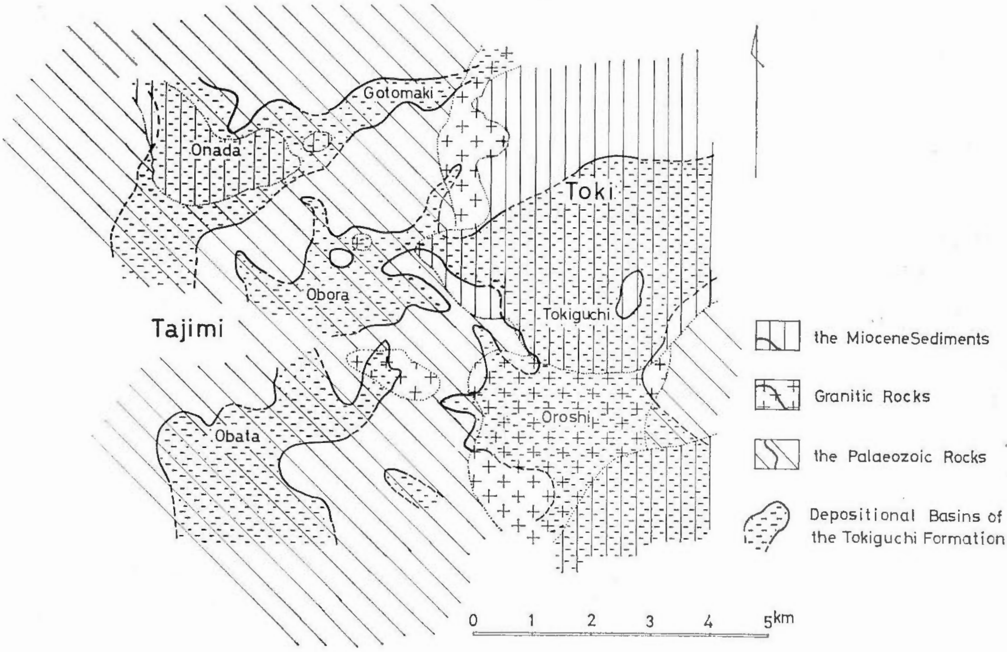



Fig. 2 Paleogeographical map of Tajimi and Toki basins

It is noted that fireclay deposits in each basin occur with distinct areal-features. For instance, large scale gaerome-clay deposits are found in Toki basin, but do not exist in Tajimi basin, with one exception of a small size deposit occurring at the northern part of Obora sub-basin. Most of kibushi-clay deposits are developed in Tajimi basin, but in Toki area they are found limitedly at marginal parts of the basin.

Moreover the so-called kaolin deposits are mainly found in Obora sub-basin and a part of Obata one. And "kaolin" deposits found in Toki basin are of small size and poor quality, and in Onada sub-basin even poor quality "kaolin" deposits are seldom found.

It appears that such differences in the occurrence of fireclay deposits between the two basins depend mainly on the difference of basement rocks between the two. In the following sections, stratigraphical sequence and geological structure of the Tokiguchi formation in each basin will be described briefly.

## III. 2 The Tokiguchi formation in Onada sub-basin

### III. 2. 1 Stratigraphy

Pre-Pliocene system in Onada area is mainly composed of Palaeozoic chert, shale and sandstone, and a part of them is overlain by tuffaceous sedimentary rocks belonging presumably to Miocene series. It has been ascertained by boring that the Miocene beds are distributed mainly at the western part of this area.

The Tokiguchi formation is distributed in a long and narrow area trending east-west. It consists largely of frequent alternations of gravel, sand and clay beds, accompanying a few lignite seams. It is observed often that these lignite seams are accompanied by kibushi-clay beds (Fig. 3).

Stratigraphical sequence of the Tokiguchi formation in this area is as follows; basal gravel bed, sand and gravel rich facies, clay rich facies, sand and granule rich facies and silt rich facies in ascending order. Either of these facies is not always uniformly developed at any part of this area (Fig. 4).

For instance, the basal gravel bed is distributed thickly at the eastern part of this area but thins out at the western part. The sand and gravel rich facies are also developed thickly at the eastern part. On the other hand, the clay rich facies are mainly distributed at the western part, and accompany a few kibushi-clay beds, which are exploited at 10 workings. The most workable bed is more than 4 meters in thickness at some places. Also at Gotomaki, the

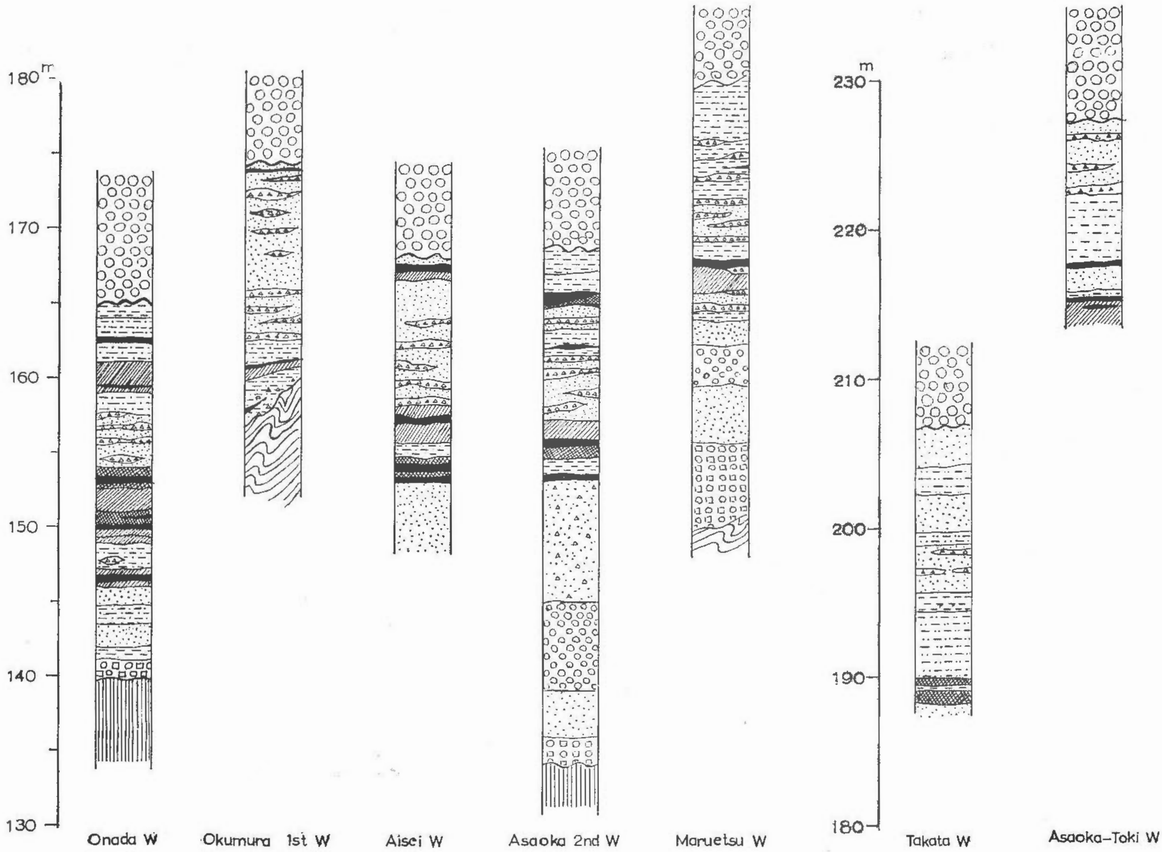



Fig. 4 Columnar section of the Tokiguchi formation in Onada area (Legend in Fig. 19)

eastern part of the area, a kibushi-clay bed is found. This bed is less than 2 meters thick and is worked at two workings. But at the central part of this area kibushi-clay bed is seldom found.

The uppermost silt rich facies may accompany a few lignite seams with kibushi-like clay beds. But these clay beds are of poor quality and they frequently thin out.

### III. 2. 2 Process of sedimentation

It seems likely that such variable occurrences of each facies in the Tokiguchi formation as described above would be mainly controlled by the basement topography. Isopach lines of the clay rich facies at the western part is shown on Fig. 5. It is noteworthy in this figure that the clay rich facies at the west side of the central buried hill develop more thickly than at the east side. These facts may suggest that at the opening of deposition of the Tokiguchi formation, a large quantity of coarse-grained materials were transported from east and were deposited at the front of the buried hill.

It seems probable that such differential deposition separated Onada sub-basin and formed the two swamps, which are favourable for deposition of lignite and kibushi-clay.

After deposition of the clay rich facies, Onada sub-basin probably subsided and was filled up mostly by rapid deposition of the sand and granule rich

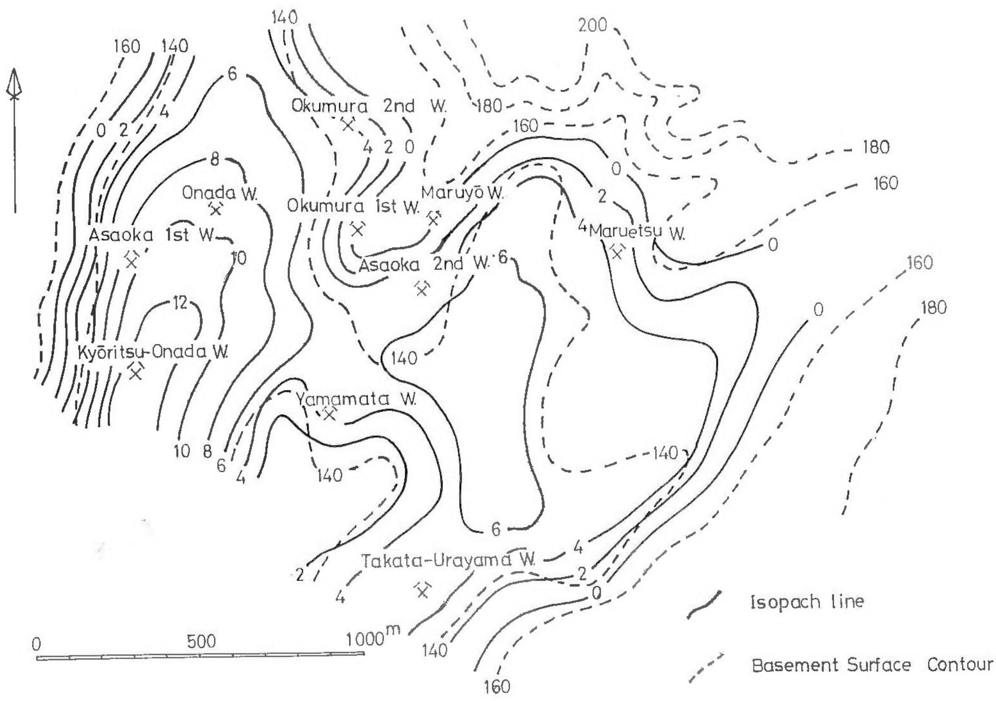



Fig. 5 Isopach map of the clay rich facies of the Tokiguchi formation in Onada area

facies. The uppermost silt rich facies would have been deposited at a shallow swamp finally. Moreover, the Toki formation was deposited in a wider area overlying the Tokiguchi formation disconformably.

### III. 2. 3 Geological structure

In Onada area, the Tokiguchi formation seems to be lying horisontally. But the results of precise leveling clarified that this formation dips monoclinaly at about 5° to southwest or south. And it is also observed frequently that this formation dips more than 10° near the barriors of the basement.

There are a few faults cutting the Pliocene formations. They are of small scale and strike in the direction from northwest to southeast along Nishibora river at the western part.

## III. 3 The Tokiguchi formation in Obata sub-basin

### III. 3. 1 Stratigraphy

The basement rocks of this area consists of Palaeozoic rocks, which are chert, shale, sandstone etc. This sub-basin is of very complex shape and its margin is complicated. Stratigraphical sequence of the Tokiguchi formation in this area is as follows; gravel and sand rich facies, clay rich facies, sand and granule rich facies and clay rich facies in ascending order. This sequence may be similar as a whole to that in Onada area (Figs. 6, 7).

Moreover basal gravel bed is also distributed locally at the southeastern part of this area, but is continuous intermittently.

The gravel and sand rich facies is distributed thickly at the southern and western parts of the area. They are composed dominantly of pale blue sand and chert breccia, and attain 10 meters in thickness. Most of the chert breccias are grey coloured and very brittle. It appears that they were altered subjecting to intense weathering before deposition. Both the upper and lower clay rich facies are accompanied by a few lignite seams, and kibushi-clay beds are well developed near Miwa workings. But at Akasaka and Iwagane workings, a kibushi-clay bed is about one meter thick, and it thins out at the marginal parts of the sub-basin. Besides kibushi-clay, a "kaolin" bed about 20 centimeters thick occurs intermittently at the southeastern part (Fig. 8).

In Obata area, some of the kibushi-clay beds are exploited at five workings, but most of them have not been worked.

And the Tokiguchi formation of this area is greatly disturbed by large scale faulting as will be described later. Therefore concrete geological evidence for interpreting the sedimentary process of the Tokiguchi formation have not yet been obtained.

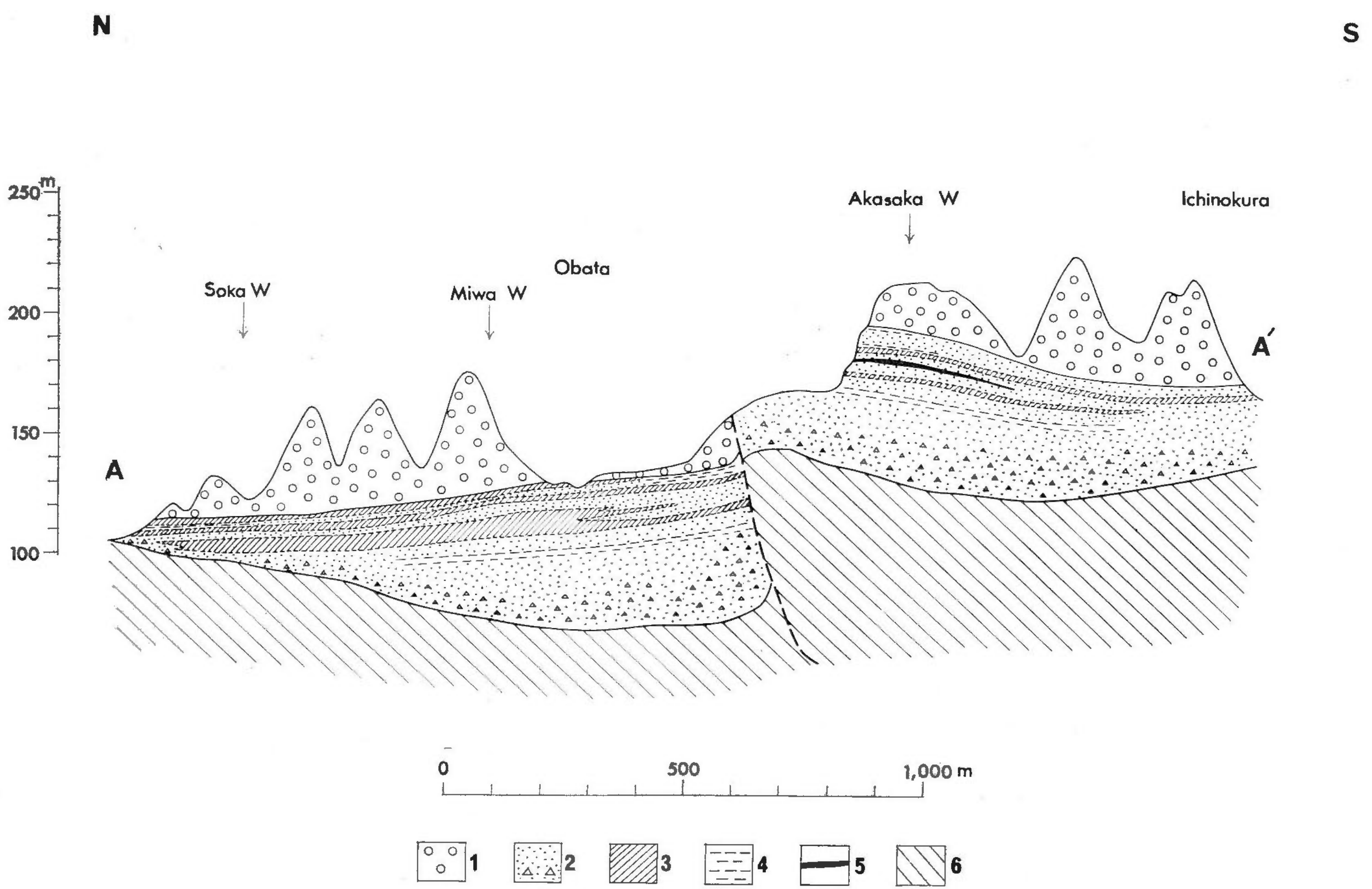



Fig. 7 Geological profile of Obata area

1 : The Toki formation.
2 : Sand and granule rich facies. 3 : Kibushi-clay bed. 4 : Clay rich facies. 5 : Tuff and/or "kaolin". } The Tokiguchi formation.
6 : The Palaeozoic rocks.

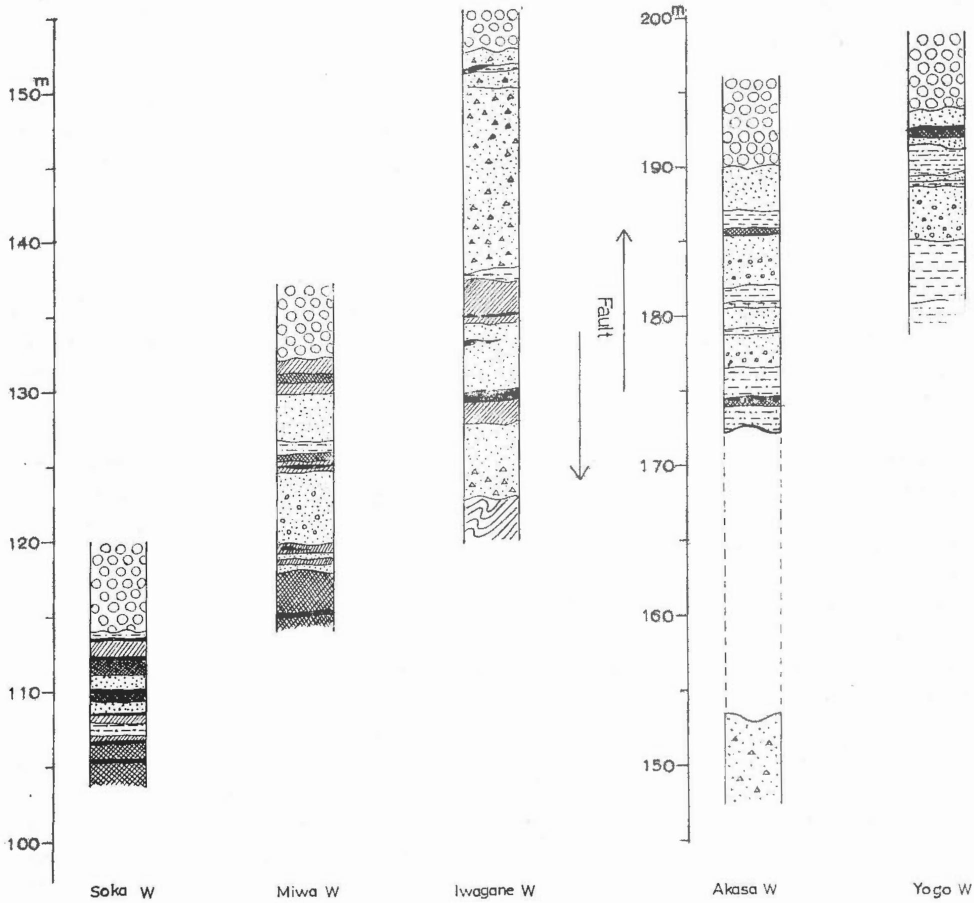



Fig. 8 Columnar sections of the Tokiguchi formation in Obata area

### III. 3. 2 Geological structure

There are two thrust faults cutting the Seto group at Obata area. One thrust strikes east-west, and the other in the direction of N. 30° W. It seems probable that the amounts of vertical displacement by faulting range from 30 to 40 meters. The Tokiguchi formation forms gently undulating structure as a whole. Especially, a gentle anticline trending N. 30° W. is found along Kasahara river. Such folding may have close relation to faulting descrived above.

### III. 4 The Tokiguchi formation in Obora sub-basin

### III. 4. 1 Stratigraphy

Obora area is one of the important clay-producing area in this district together with Toki area. A few kinds of fireclays, which are kibushi-clay, gaerome-clay and the so-called kaolin, are produced in this area. Perhaps about 70,000 tons of kibushi-clay, 10,000 tons of "kaolin" and a small quantity of gaerome-clay are mined annually.

The basement rocks are mostly of Palaeozoic in age. Besides, it appears that a small stock of granitic rocks is distributed near Yamaso workings at the northern part of this area, overlain by the Tokiguchi formation. As good evidence in support of the above assumtion, the following facts are observed; gaerome-clay presumably derived from granitic rock is distributed at only Yamaso workings, and the Palaeozoic rocks surrounding this workings were subjected to thermal metamorphism (Figs. 9, 10).

The stratigraphical sequence of the Tokiguchi formation in Obora area are generalized as follows; gaerome-clay facies, chert gravel bed, sand and gravel rich facies, clay rich facies, sand and granule facies and silt rich facies in ascending order. The sequence, with the exception of the lower two facies which are distributed locally at the northern part of this area, can be correlated easily with those in Onada and Obata areas. But at the north-eastern part, only the upper two facies of the sequence occur. The thickness of the Tokiguchi formation usually ranges from 20 to 30 meters in this area (Fig. 11, 12).

Gaerome-clay deposits near Yamaso workings extend about 200 meters in the east-west direction and about 100 meters in the north-south direction, less than 10 meters thickness. In some places, it is observed that thin silt beds are intercalated cross-laminating in the gaerome-clay deposits. Chert gravel bed less than 8 meters thick is also distributed locally, overlapping the gaerome-clay facies. It consists mainly of round gravels of chert from 2 to 10 centimeters in diameter.

It appears that the so-called kaolin occurs at two preferred stratigraphical positions in the Tokiguchi formation. The lower one occurs beneath the chert gravel bed, forming thin beds less than 10 centimeters in thickness. But its distribution is restricted to the northern part of Obora area. Commercially workable "kaolin" deposits are of the upper position, which generally corresponds to the lower part of the silt rich facies. These "kaolin" beds are generally lenticular in shape, usually from 20 to 50 centimeters in thickness. In the neighbourhood of Shinmei path, the north-eastern part of Obora area, it is best developped, attaining a thickness of 2 meters in some places. The so-called kaolin can be distinguished usually by its white coloured appearance and homogeneity from kibushi-clay and others. "Kaolin" bed usually has sharp contacts with enclosing sediments, but it was observed at some places that it grades into crystal tuff. Moreover, microscopic observation of "kaolin" shows that volcanic glass shards lie in isotropic matrix (Plate III–1, 2). These facts suggest that "kaolin" was formed authigeneously from volcanic materials.

In Obora area, kibushi-clay occurs at three preferred stratigraphical horizons in the Tokiguchi formation. The lowest kibushi-clay bed occurs at similar horizon as the gaerome-clay facies, but it appears that its occurrence is restricted to a narrow range near Obora 2nd workings.

The middle horizon of kibushi-clay bed, which is the most workable in this area, is developed in the clay rich facies. This bed is distributed in the Obora sub-basin excepting the northeastern part, attaining a thickness of 3 meters in some places. And it tends to thin out westwards. The uppermost kibushi-clay bed, which occurs in the silt rich facies, is developed only at the north-eastern and western parts. Kibushi-clay of this horizon usually contains some

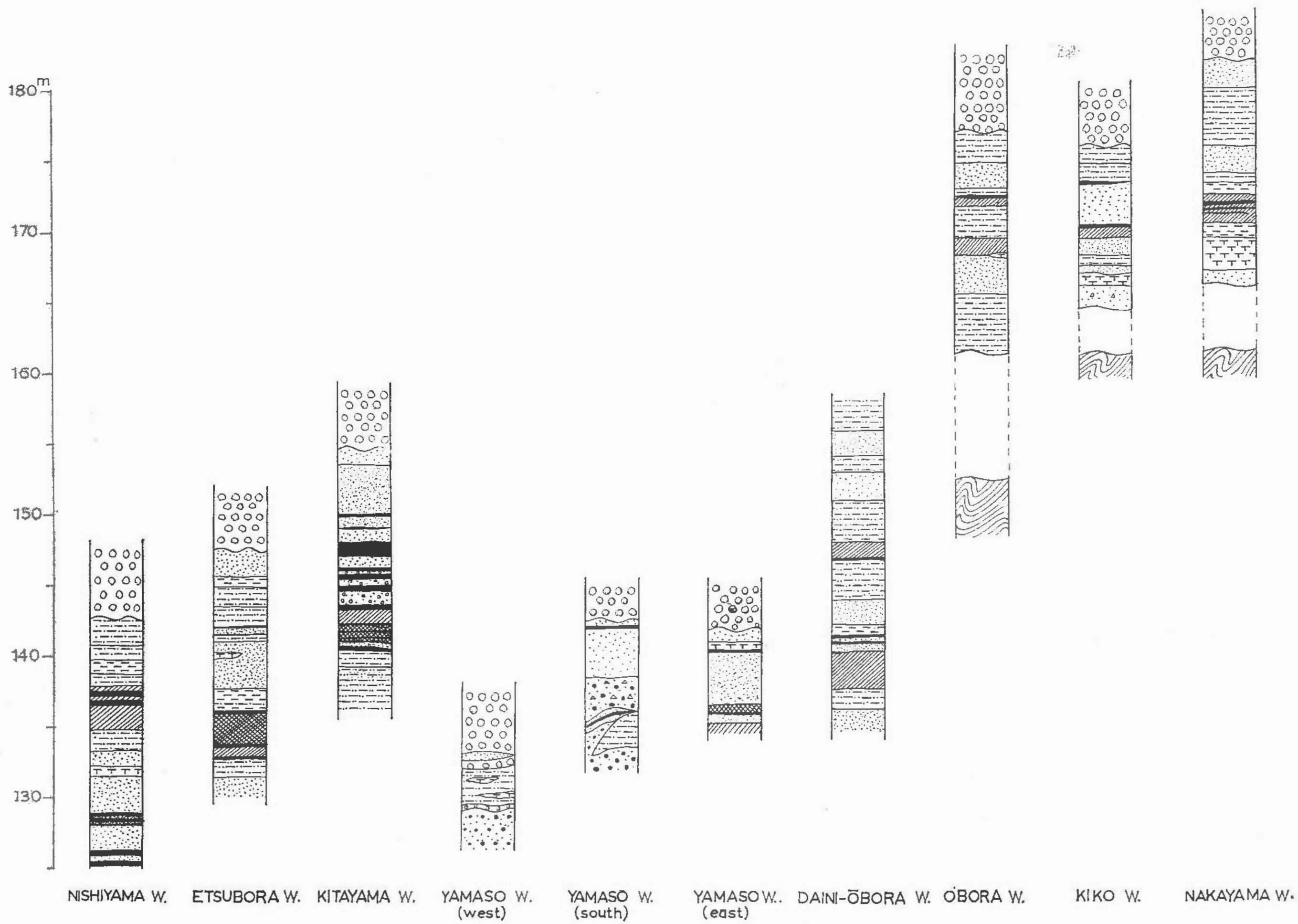



Fig. 11 Columnar sections of the Tokiguchi formation at the northern part of Obora area (Legend in Fig. 19)

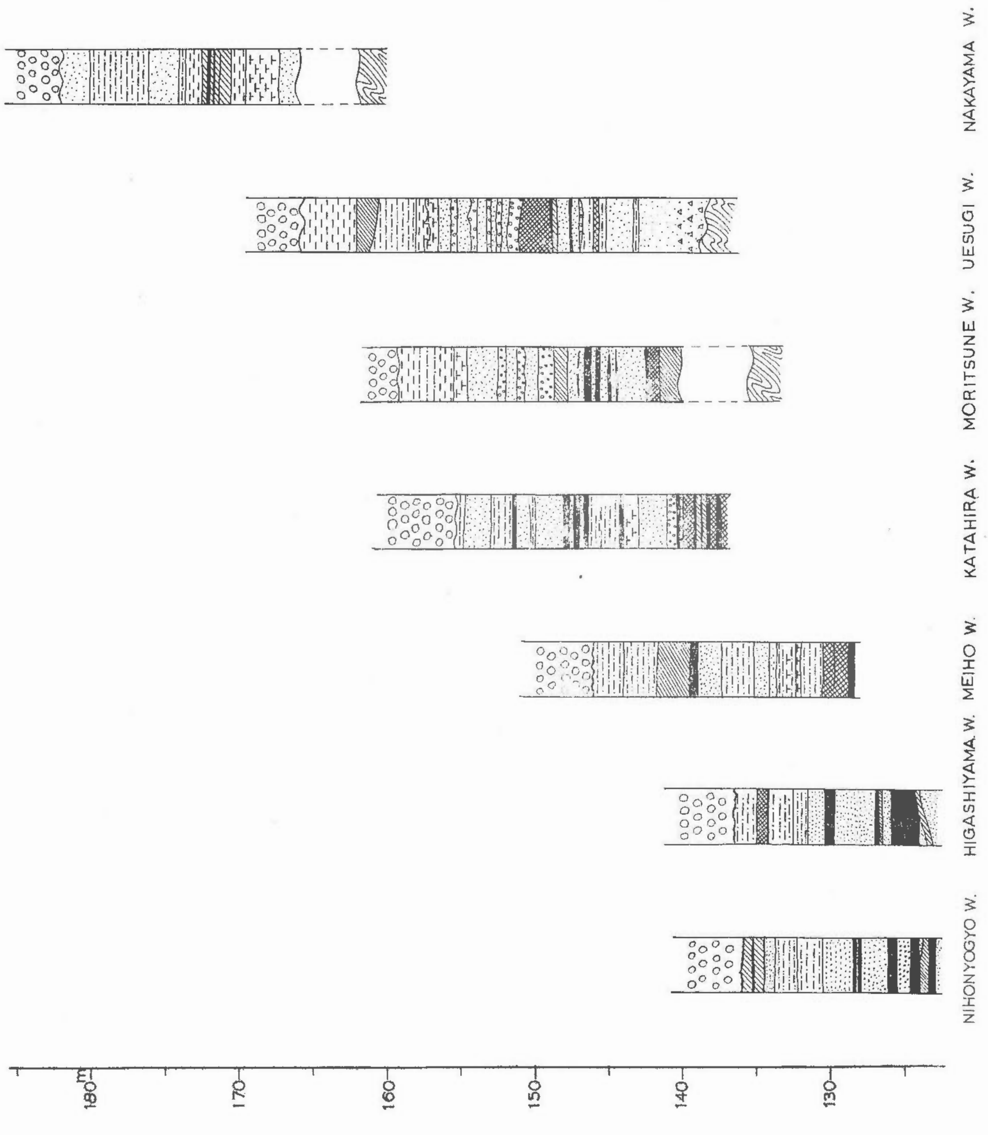



**Fig. 12 Columnar sections of the Tokiguchi formation at the central part of Obora area**

sand grains, and many are of poorer quality than those of other horizons.

Workable miscellaneous clays also occur in the Tokiguchi formation. They are usually dark or pale blue and mostly of SK 26 to 30 refractoriness. And so they are used as ceramic raw materials and binder of molden sands.

### III. 4. 2 Process of sedimentation

Fig. 13 is the structure contour map referring to the basement surface of the time of deposition of the Tokiguchi formation. The distribution extents of the upper two kibushi-clay beds and the gaerome-clay facies are also shown on the

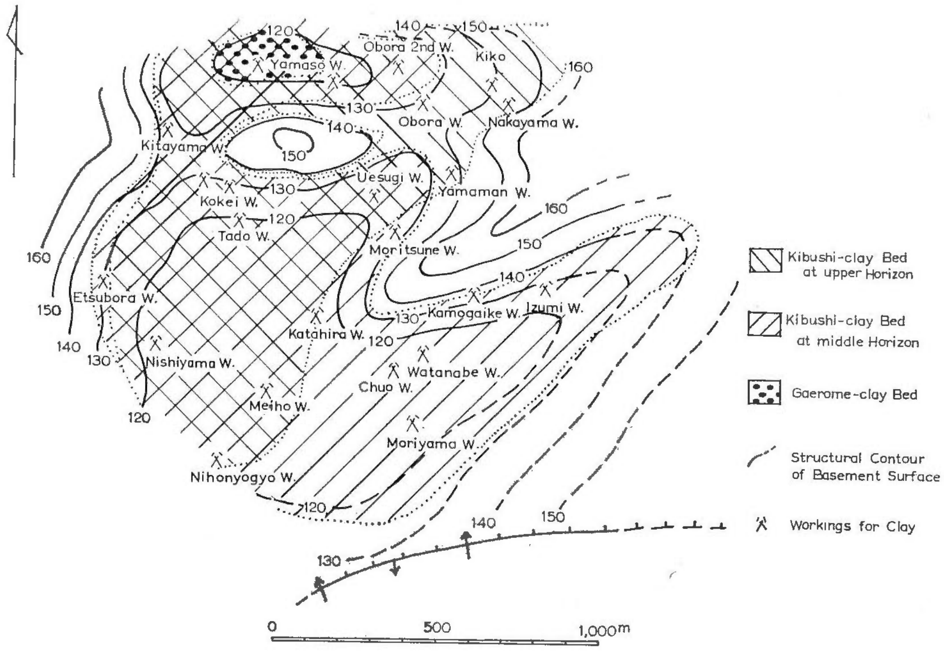



Fig. 13 Sedimentary process of the fireclay beds at Obora sub-basin

same map. This map suggests that most of the sediments, belonging to the Tokiguchi formation, were supplied to this sub-basin from east and northeast through a few channels. At that time, there may have been an oval shaped depression at the northern part, presumably formed on basement granite which was easily eroded.

Sedimentation of the Tokiguchi formation in Obora sub-basin began with the deposition of the gaerome-clay facies and others at the depression mentioned above. It was followed by deposition of the chert gravel bed. It appears that rapid subsidence in this district gave rise to deposition of this gravel bed. At that time, the depositional area was enlarged probably to almost whole part of Obora sub-basin besides the northeastern part. Successively the clay rich facies, with lignite seams and kibushi-clay beds, were deposited under a quiet environment. Moreover they were followed by deposition of abundant coarse-grained materials, extending over the whole Obora sub-basin. This sub-basin was nearly filled by a sequence of deposition as stated above. But as the result of differential deposition in the sub-basin, it is assumed that a few swamps were formed at the northeastern and western parts. The above assumption may be proved by limited distribution of the upper horizon of kibushi-clay bed.

### III. 4. 3 Geological structure

In this area, the Tokiguchi formation shows a monoclinal structure dipping generally to southwest about 5°. The distribution of this formation is restricted to the northern side of east-west trending fault, which is likely to be a high angle reverse fault from many geological evidences. In addition, the Toki formation, underlain by the Tokiguchi formation, is cut by this east-west trending fault. These facts suggest that this fault began to be formed at the pre-Pliocene epoch and its movement succeeded after deposition of the Seto group. At the south-eastern part of Obora area, the Tokiguchi formation appears to dip northwest being subject to displacement of this fault.

## III. 5 The Tokiguchi formation in Toki basin

### III. 5. 1 Stratigraphy

The survey by the writer confirmed that Toki basin is at least 3 km in the east-west direction and 5 km in the north-south direction in dimensions. In addition, this basin seems to be opened southwards and northeastwards. Therefore it can be said that Toki basin is far larger than either of three sub-basins described previously.

Most of the basement rocks in this basin are granitic rocks and Miocene sediments, which are called the Toki coal-bearing formation and the Mizunami group. The Tokiguchi formation in Toki basin is generally divided into two parts, upper and lower. The lower part of this formation is composed mainly of gaerome-clay and pale blue fine sand. On the other hand, the upper part consists of arkose sand rich facies and clay rich facies. It is observed that the clay rich facies intercalate a few beds of tuff and tuffaceous clay. Moreover, it is notable that chert gravel bed is developed locally at the horizon between the upper part and the lower one (Figs. 14, 15).

Gaerome-clay deposits are distributed at the central part of this area, attaining a thickness of more than 20 meters. It is observed frequently that thin beds of light green micaceous-sand are intercalated in the gaerome-clay deposits. They are not homogeneous in sorting grade, and usually include 5 to 30 per cent clay. This deposits are mined at more than 10 workings. The gaerome-clay facies tend to grade into fine sand rich facies towards the margin of basin. This fine sand facies usually includes considerable amount of clay fraction, and can be used as ceramic raw material as well as gaerome-clay.

The chert gravel bed consists mostly of round gravels of chert, which are from 1 to 10 centimeters in diameter, and coarse sand matrix. It appears that this bed occurs in a wide channel of north-south direction where the gaerome-clay facies had presumablly been washed out by rapid influx of chert gravels. The maximum thickness of this gravel bed is about 10 meters. A similar gravel bed is distributed near Nishiyama workings at the southwestern part of the basin. But its distribution extent has not been confirmed.

Overlapping conformably these facies stated above, clay rich facies and arkose sand rich facies are distributed over the whole basin (Fig. 16). Their total thickness ranges from 10 to 15 meters. A few beds of tuff and tuffaceous clay also occur in the clay rich facies. But with one or two exceptions, it seems that good quality "kaolin" does not occur in Toki area as in the case of Obora area.

Considering from their stratigraphical sequence and occurring level, it is highly possible that the clay rich facies are correlated with the silt rich facies in Obora area (Fig. 17). It may be assumed that Toki basin was connected

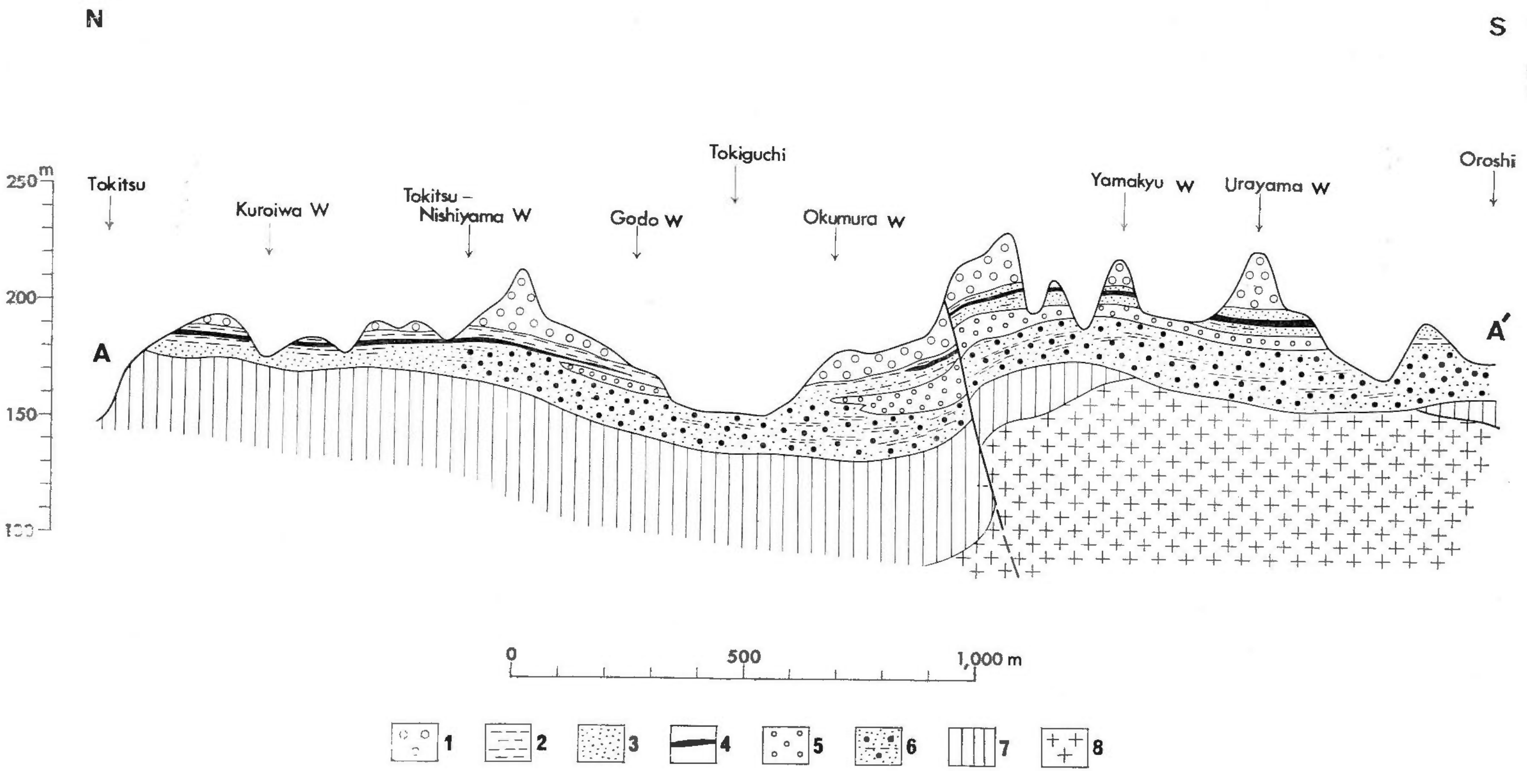



Fig. 15 Geological profile of Toki area

1: The Toki formation.
2: Clay rich facies.
3: Sand rich facies.
4: Tuff and/or "kaolin".
5: Chert gravel bed.
6: Gaerome-clay facies.
7: The Miocene series.
8: Granitic rocks.

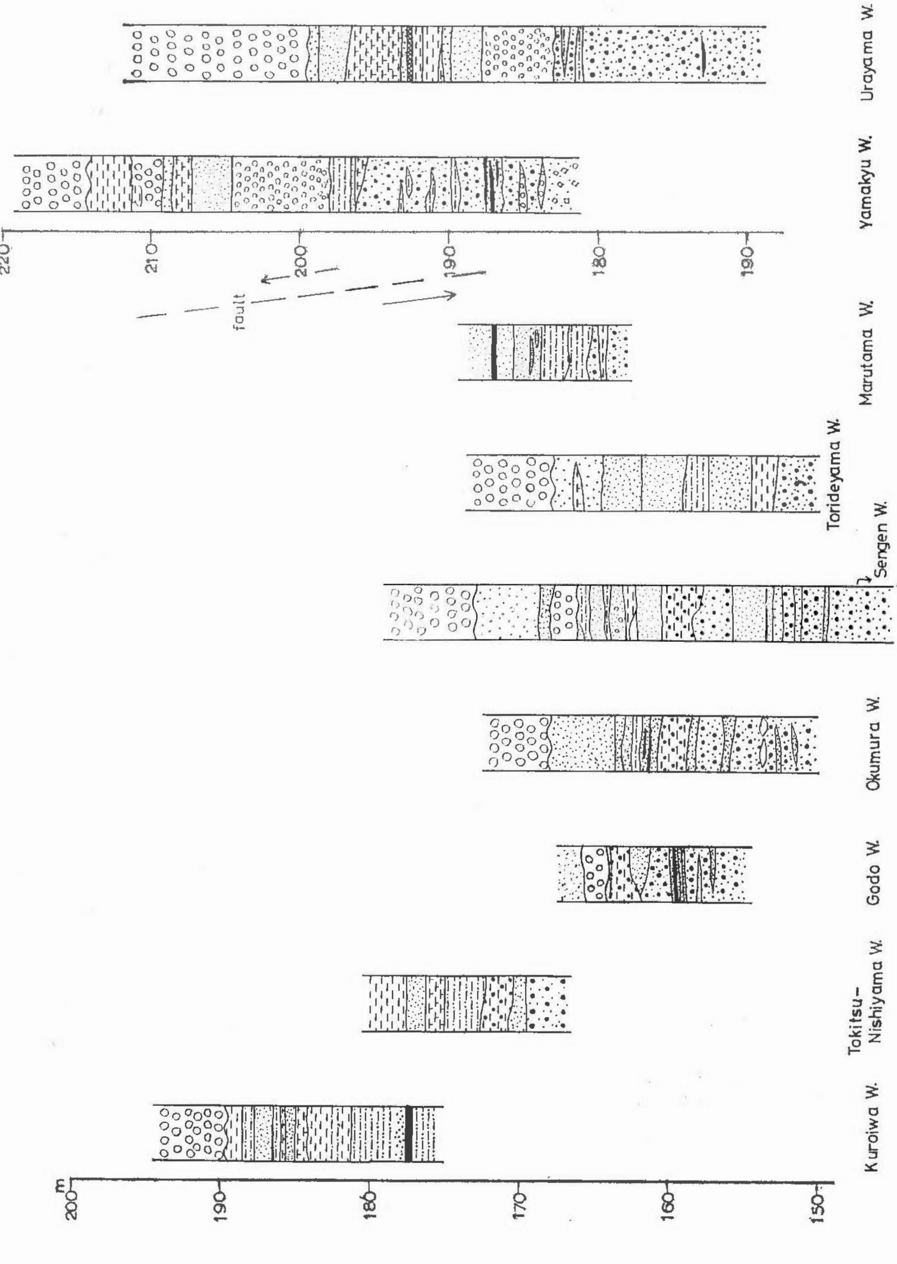



Fig. 16 Columnar sections of the Tokiguchi formation in Toki area (1)

with Obora sub-basin by a narrow channel at the time of deposition of the clay rich facies. In addition, kibushi-clay deposits are also developed in the same facies at the southwestern and southeastern marginal areas of Toki basin.

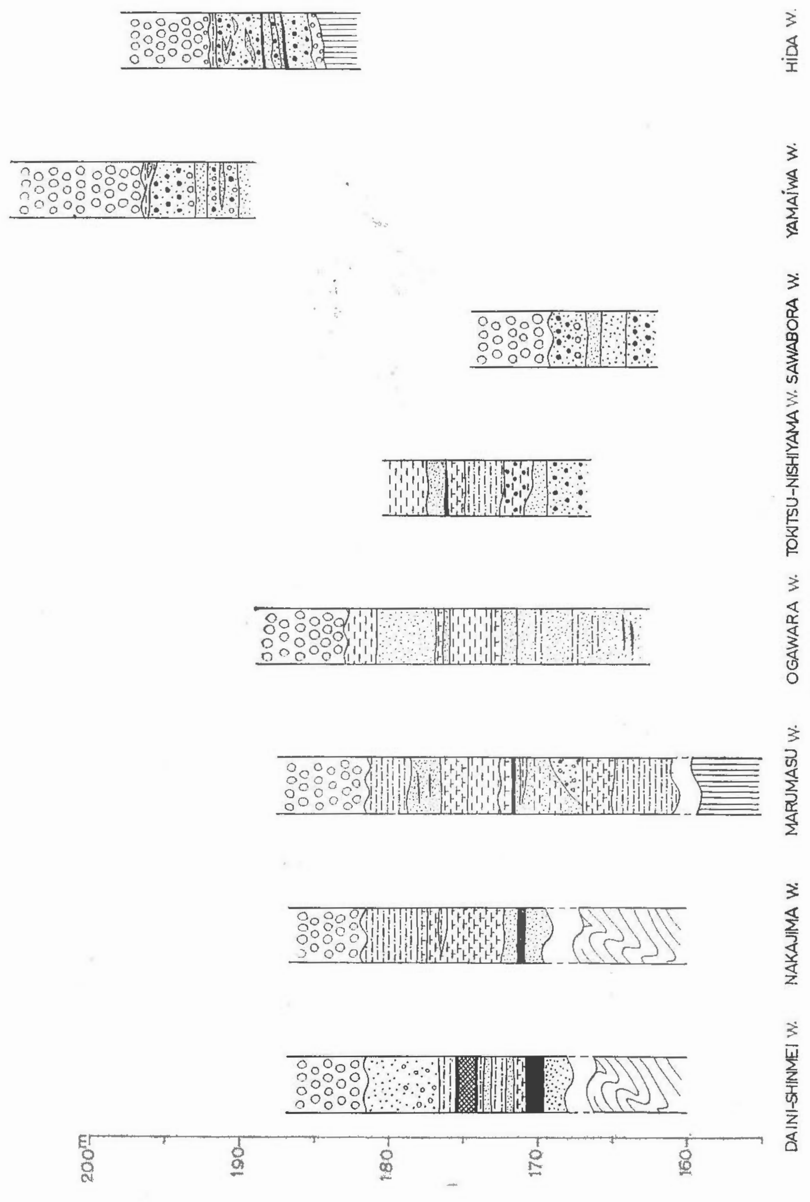



Fig. 17 Columnar sections of the Tokiguchi formation in Toki area (2)

## III. 5. 2 Process of sedimentation

Toki basin is too extensive to grasp precisely its basement topography. But it is possible to infer the basement topography by the occurrence of the Tokiguchi formation and some boring data.

On Fig. 18, marginal lines of the basin and isopach lines of the lower part of the Tokiguchi formation are shown. These isopach lines suggest that there

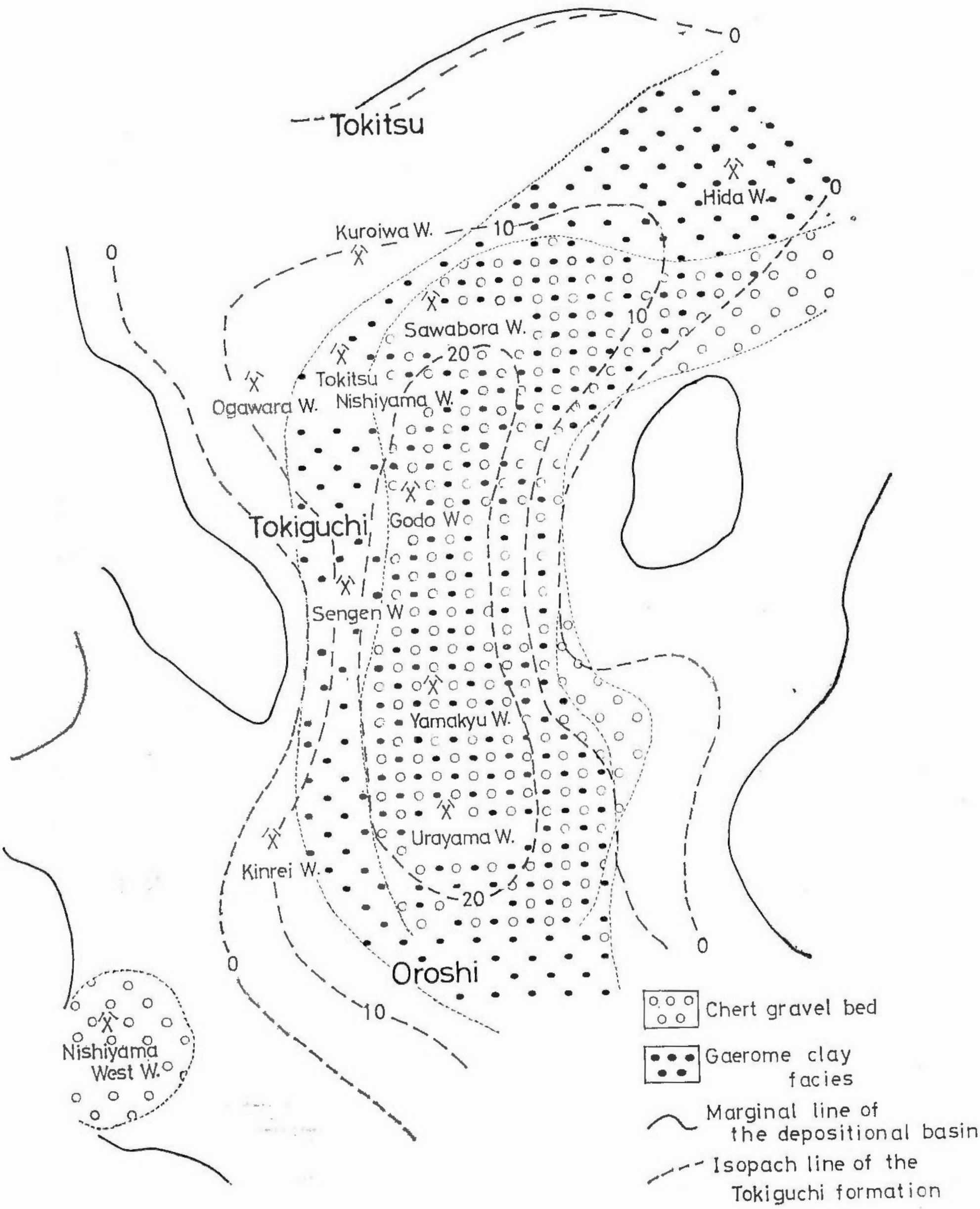



Fig. 18 Sedimentary process of the Tokiguchi formation at Toki basin

is a wide depression trending in north-south direction at the central part of Toki basin. Distribution of the gaerome-clay facies seems to be restricted to this depression.

From many geological evidences, sedimentary process of the Tokiguchi formation is summarized as follows. First of all, gaerome-clay facies and pale bule fine sand facies were deposited simultaneously, the former at the central depression and the latter at the marginal parts of this basin respectively. Succeedingly, rapid subsidence gave rise to local deposition of the chert gravel bed. It was followed by slow deposition of the clay rich facies and the arkose sand rich facies. At that time, kibushi-clay beds were deposited at some marginal areas in the southwestern and southeastern parts.

And it is noteworthy that a few beds of tuff and tuffaceous clay occur at upper position than the horizon of the chert gravel bed. This fact suggests that volcanism began at nearly the same time with subsidence, which gave rise to deposition of the chert gravel bed.

### III. 5. 3 Geological structure

In general, the Tokiguchi formation dips gently southwards in Toki area. It undulates subjecting to movement of a fault, which runs from east to west through the central part. It appears that this fault was formed by northward thrust movement. It may be correlated with the reverse fault setting the southern bound of Obora sub-basin. Many geological evidences suggest that this fault commenced to move before deposition of the Tokiguchi formation. It is probable that it is included in Yamada fault zone, which was named by MATSUZAWA and UEMURA (1964).

## IV. Consideration of Sedimentary Process of the Tokiguchi Formation

### IV. 1 Correlation

In this section, sedimentary process of the Tokiguchi formation will be summarized based on geological data mentioned above.

Stratigraphical sequence of the Tokiguchi formation at each basin can be generalized as Fig. 19. The Tokiguchi formation shows a very similar sequence in either of the three sub-basins in Tajimi area, with a few exceptions. On the other hand, it is observed that there are distinct differences in stratigraphical sequence between the Tokiguchi formation of Tajime area and that of Toki area. But when the columnar section of Toki basin is compared with that of the adjacent Obora sub-basin, it may be found that there are some features common between the two. These features are as follows.

(1) In either area gaerome-clay facies, which are different in scale, always occur at the basal part of the Tokiguchi formation.

(2) In either area, the chert gravel bed occurs underlain by the gaerome-clay facies. It seems probable that rapid subsidence in this district gave rise to deposition of these gravel beds. At that time, the depositional area was enlarged more remarkably at Obora sub-basin than at Toki basin (Figs. 13, 18).

(3) In Toki basin, a few beds of tuff and tuffaceous clay occur at upper horizon than the chert gravel bed. In Obora sub-basin, a major "kaolin" bed occurs at a far upper position than the chart gravel bed. But at the northern part, a thin bed of "kaolin" occurs beneath the chert gravel bed. From some

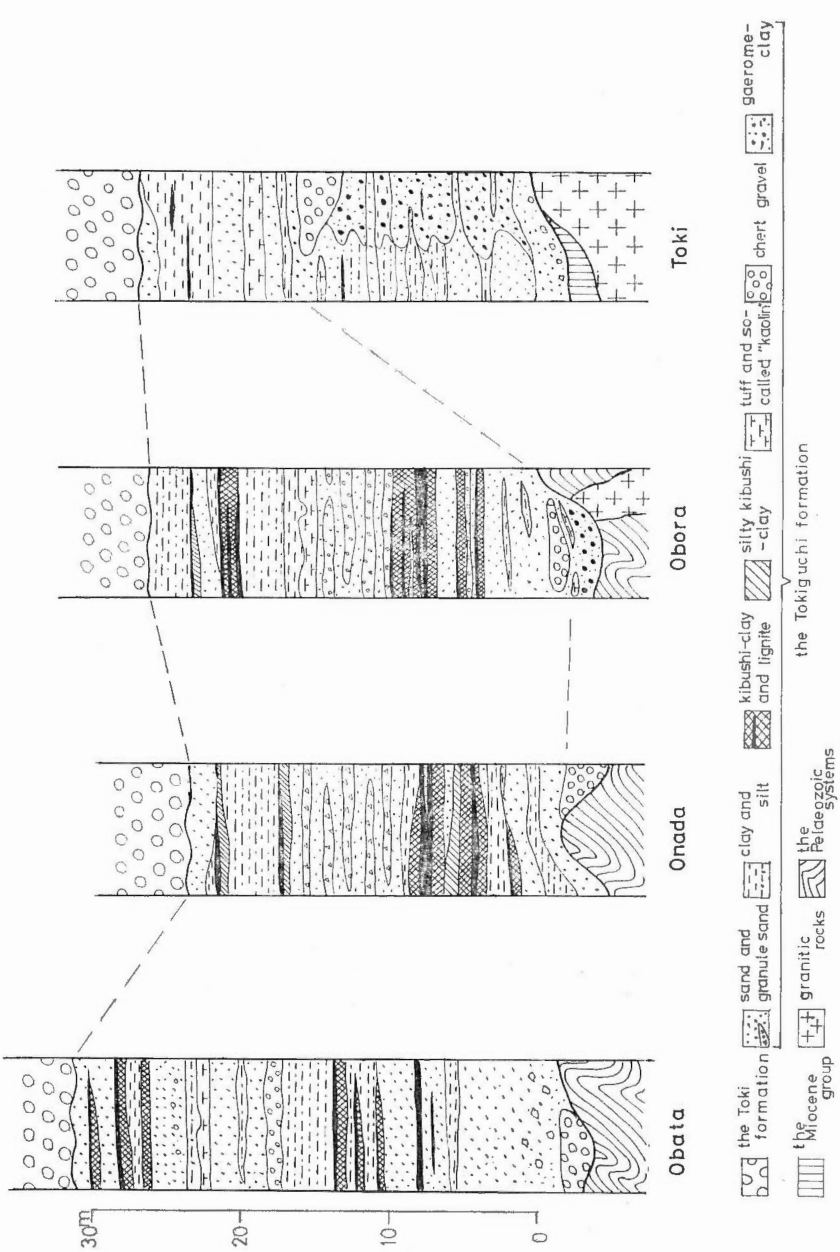



Fig. 19 Generalized columnar sections of the Tokiguchi formation

evidences as stated previously, it is inferred that "kaolin" was formed diagenetically from tuffaceous material.

From these facts, it is possible that the chert gravel beds may have deposited simultaneously at both basins. Similar gravel beds are distributed locally at the basal part in both Onada and Obata sub-basins.

## IV. 2 Sedimentary process

From the results of the correlation mentioned above, it is concluded that the Pliocene series were deposited in the following process (Fig. 20).

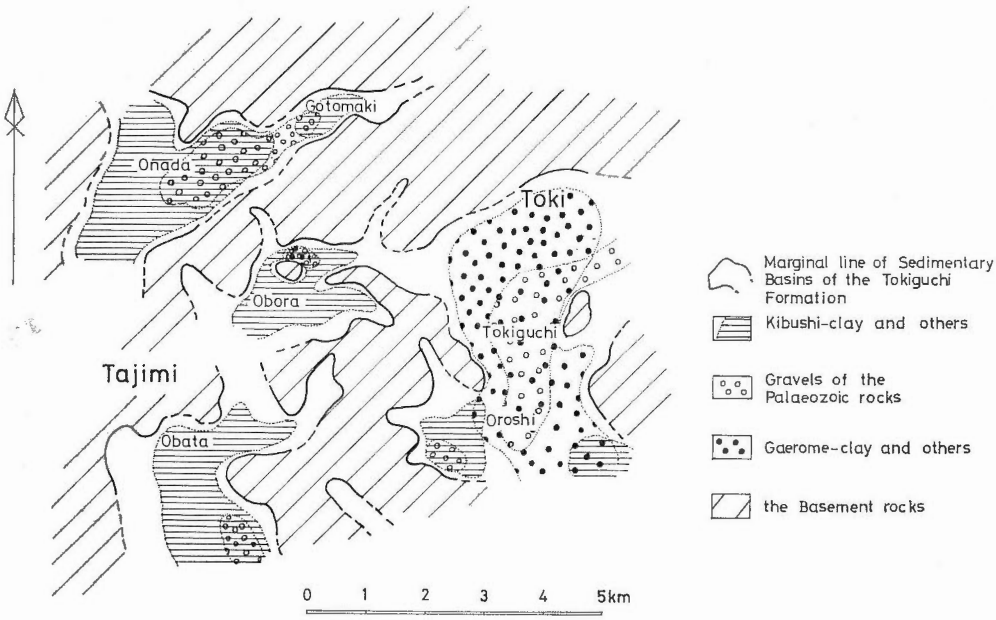



Fig. 20 Sedimentary process of the Tokiguchi formation in Tajimi-Toki district

(1) The Pliocene sedimentation originated with deposition of thick gaerome-clay facies and fine sand facies at Toki basin. But most of Tajimi basin still remained without deposition. As an exception, gaerome-clay facies were deposited only at the northern part of Obora sub-basin.

(2) Successively, regional subsidence gave rise to deposition of chert gravels at each basin. At that time, depositional area of Tajimi basin was enlarged rapidly, and abundant clastic materials began to be deposited into each sub-basin. And a few lignite seams and kibushi-clay beds were formed at Tajimi basin, probably under the lacustrine or swamp environment. Additionally, it seems that volcanic activity took place in an unknown area at nearly the same time with deposition of the chert gravel beds and began to supply tuffaceous materials into some basins.

(3) Next, areal subsidence caused deposition of a sequence of granule, sand and silt beds with 15 to 25 meters thick at Tajimi basin. On the other hand, at Toki basin, a little subsidence gave rise to deposition of a thin bed consisting mainly of tuffaceous clay and arkose sand.

(4) After the deposition of the Tokiguchi formation, an extensive subsidence took place and caused the deposition of flood plain sediments that

consists mainly of round gravels of chert, quartz porphyry and so on. These sediments are named the Toki formation. This formation was deposited overspreading the whole area of this clay-producing region, and is 20 to 50 meters in thickness. Therefore, it is believed that this formation was formed over peneplainlike hilly-region, specific heights of which were comparatively small.

### IV. 3 The relationship between the basement rocks and sedimentary features of the Tokiguchi formation

The differences in stratigraphical sequence and sedimentary process of the Tokiguchi formation between Tajimi and Toki basins seem to depend mainly on the difference of basement geology between the two. For instance, the distribution of granitic rocks in the basement seems to be the best way of explaining deposition of abundant gaerome-clay at Toki basin.

Moreover, it can be said that each depositional basin is characterized by the basement geology. For example, all the sub-basins in Tajimi area, the basement of which is composed mainly of the Palaeozoic rocks, are of very complex shape and very undulated. These facts suggest that dense drainages were developed at Tajimi area, and such circumstances would have promoted heavy vegetation in this area.

On the other hand, the basement rocks of Toki area, which consist dominantly of granitic rocks and the Miocene sediments, were probably susceptible to weathering and denudation. And so Toki basin is characterized by comparatively extensive and gently undulated features. Gaerome-clay facies seems to be the sediments resulting from the rapid dumping type of deposition of degraded granitic rocks. The occurrences of kibushi-clays at Tajimi basin also have close relation to the basement geology, because heavy vegetation would give rise to formation of lignite seams. Moreover, it may be said that complicated reliefs were favourable for formation of swamps, which were suitable for the deposition of lignite seams and kibushi-clay beds (Figs. 5, 13).

Next, the difference of the development of the so-called kaolin deposits between Obora sub-basin and Toki basin may be explained as follows; slow sedimentation of the upper part of Tokiguchi formation at Toki basin gave rise to formation of tuffaceous clay beds that contain many clastic materials.

On the other hand, rapid deposition of clastic materials may have caused development of pure tuff beds at Obora sub-basin. These facts suggest that the formation of "kaolin" of good quality was probably influenced by both clastic impurities and lake acidity.

## V. Tectonic Movement after Deposition of the Pliocene Group

The Tokiguchi formation in this district shows gently undulated structure affected by faulting and tilting as stated previously. But the displacement of the Tokiguchi formation by these tectonic movements generally seems to be the same as that of the Toki formation. In other words, it can be said that the Tokiguchi formation was affected with few tectonic movement throughout the period of its sedimentation. Moreover, it was mentioned previously that the Toki formation was deposited over extensive and peneplain-like hilly-region. These facts indicate that the structure of the basal plain of the Toki formation may generally reflect tectonic movements after deposition of the Pliocene Seto group. The structural contours of the basal plain of the Toki formation are shown on Fig. 21a, and main faults and folding axes deduced from Fig. 21a are shown on Fig. 21b.

As these figures show, it can be said that main geological structures in the

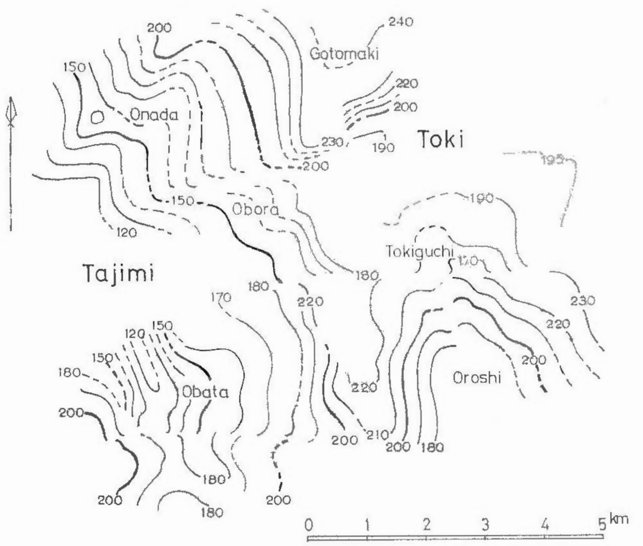



Fig. 21a Structure contour map of the basal plane of the Toki formation

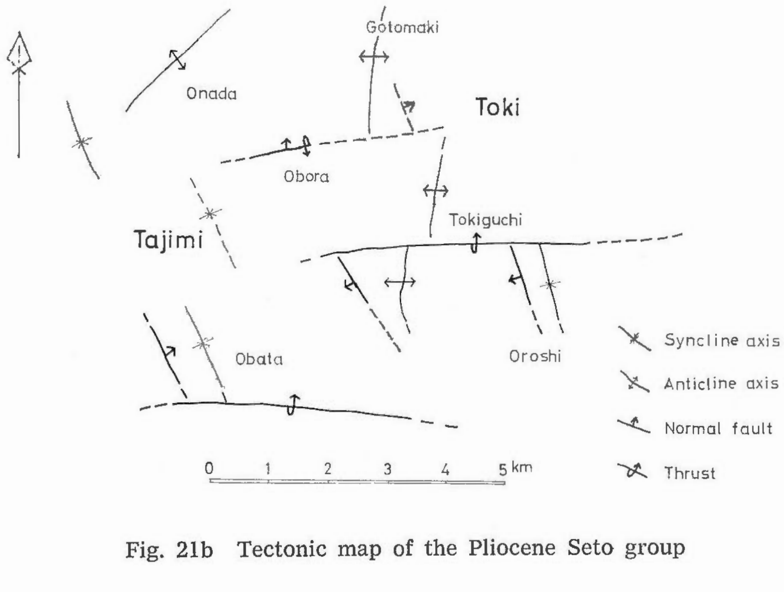



Fig. 21b Tectonic map of the Pliocene Seto group

Pliocene Seto group are the folding structures, axes of which are of north-northwest or north to south trend, and a few thrusts of east-west trend. These folding structures are generally gentle, and their axes seem to be cut by the east-west faults and displaced 1 or 2 kilometers in east-west direction. Relating to the east-west faults, the existence of at least three thrusts were recognized.

The northernmost fault seems to have been formed by stress from north, and the southern two by stress from south. At present, Toki river seems to flow along the middle graben formed by these movements. The middle thrust of these three seems to have been formed before the Pliocene sedimentation. It can be said that it is a trail of movement of Yamada fault group at the Miocene age reported by MATSUZAWA and UEMURA (1954).

## VI. Clays and Clay Minerals

### VI. 1 Macroscopic and microscopic observation

As stated previously, three kinds of fireclays, which are kibushi-clay, gaerome-clay and the so-called kaolin, occur in the Tokiguchi formation of this district. Besides, most of the pale blue silt and fine sand in this formation can probably be used as ceramic raw materials, also. And so the name miscellaneous clays will be used for these silt and fine sand in this paper.

#### VI. 1. 1 Kibushi-clay

Most of kibushi-clays are blackish brown to dark brown and contain a few lignite fragments. Good quality kibushi-clay contains few sand grains, and is highly plastic and frequently traversed by slickensided surfaces that are randomly oriented. Microscopic observation shows that most of kibushi-clays are characterized by dendritic, mesh or fibrous fabrics, the relatively strong birefringence of which is probably caused by oriented arrangement of kaolin minerals (Plate I–1, 2). Moreover it is observed that these fabrics are developed only at the pale brown parts in thin sections. These facts show that kaolin minerals in kibushi-clay grew larger to some extent after their deposition and their growth was presumably influenced by organic materials.

These fabrics are never observed in grey kibushi-clay and miscellaneous clays, which contain few organic meterials.

#### VI. 1. 2 Gaerome-clay

Gaerome-clay is composed mainly of grey clay and quartz grains, with fine grains of feldspar and mica minerals. Its clay fraction is estimated roughly to be 5 to 30 percent by washing tests (Table 2). As results of microscopic observation, it is seen that ill-sorted angular grains of quartz of more than 1 milimeter in diameter are set in a matrix containing flaky fragments of altered mica, fine quartz and clay minerals. Feldspar seems to be subordinate (Plate II-2).

And in this paper, the use of the name gaerome-clay is restricted in secondary clay derived from weathered granite and does not mean residual kaolin.

Table 2 Results of separation test by washing on gaerome-clay and others at Toki area

| No. | Specimen: Locality | Specimen: Name | Classified Fraction (wt. %): coarser (>50μ) | intermediate (10μ< <50μ) | clay (<10μ) |
|---|---|---|---|---|---|
| T-13 | Nishiyama W. (e) | pale blue silt | 43 | 22 | 35 |
| T-20 | Kinrei W. | pale blue sand | 46 | 36 | 18 |
| T-26 | Urayama W. | pale blue silt | 56 | 15 | 29 |
| T-27 | Urayama W. | gaerome-clay (sand rich) | 87.5 | 8 | 4.5 |
| T-34 | Yamakyu W. | gaerome-clay | 80.5 | 12 | 7.5 |
| T-37 | Sengen W. | gaerome-clay | 17 | 55 | 28 |
| T-38 | Sengen W. | gaerome-clay (sand rich) | 79 | 12 | 9 |
| T-39 | Sengen W. | gaerome-clay (sand rich) | 75 | 19 | 6 |
| T-41 | Torideyama W. | gaerome-clay (sand rich) | 74 | 15.5 | 10.5 |
| T-45 | Godo W. | gaerome-clay (sand rich) | 74 | 16.5 | 9.5 |
| T-46 | Godo W. | gaerome-clay (sand rich) | 42 | 42 | 16 |
| T-54 | Tokitsu-Nishiyama W. | gaerome-clay (sand rich) | 41 | 41 | 18 |
| T-59 | Sawabora W. | gaerome-clay (sand rich) | 79 | 15 | 6 |
| T-61 | Yamaiwa W. | gaerome-clay (sand rich) | 75 | 14.5 | 10.5 |
| T-64 | Hida W. | gaerome-clay (sand rich) | 75 | 17 | 8 |
| T-77 | Yamagami | gaerome-clay (sand rich) | 74 | 7 | 19 |

### VI. 1. 3 "Kaolin"

It is inferred from the geological occurrences that white clay called "kaolin" was originated from volcanic materials. "Kaolin" can be distinguished by its distinct whiteness from the other clays. But some "kaolin" occur accompanied by lignite seam. They are of brownish colour, and thus are apt to be mistaken with kibushi-clay. "Kaolin" of good quality is generally micro-grained, compact and somewhat glossy.

Microscopic observations of "kaolin" samples show that prismatic or feather-like patches of kaolin minerals and quartz particals are scattered in apparently isotropic groundmass, which consists of micro-grained materials preserving the original shape of arcuate or ring-like glass shards (Plate III-1, 2).

Some patches of kaolin minerals are feather shaped books, and appear to be kaolinized mica fragments (Plate IV-1). These books are variable in size and scattered at random. It is probable that they were formed from mica by in situ alteration.

Many observations stated above may prove that the so-called kaolin was derived from volcanic materials, which were probably rhyolitic.

### VI. 2 Clay minerals

In this study, about 400 samples of fireclay were investigated with special reference to their mineral compositions by X-ray analysis. As results of these investigations, it was known that kibushi-clay, gaerome-clay and the so-called kaolin all contain kaolin mineral as the main clay component.

But kaolin mineral species in each fireclay, occurrence of which is not similar, are different with each other. And so, in order to clarify the difference in kaolin mineral species contained in each fireclay, differencial thermal analysis, thermogravimetric analysis and electrom microscopic observations were also carried out.

#### VI. 2. 1 X-ray analysis

**Method**

Powder samples for X-ray analysis were prepared as follows. Kibushi-clay and "kaolin" were dried in air and ground without treatment, but gaerome-clay was always washed and separated into three fractions, which are coarse, medium and fine. Only the finest fraction of the three was used for this analysis. In order to make semi-oriented aggregate, a fixed amount (0.05 g) of each powder sample on a slide-glass was kneaded with distilled water and dried. X-ray analysis of prepared samples were usually carried out under the following conditions, using the X-ray diffractometer made by Rigaku-denki Co., Ltd.

Target: Cu, Filter: Ni, Voltage: 30 KV, Current: 15 mA, Scale Fact. 8, Mult.: I, Time Const.: 4, Scan. Sp.: 2°/min., Chart Sp.: 2 cm/min.

**Results and discussions**

At least three X-ray diffraction patterns were made for each sample as follows:

1. Untreated semi-orientated aggregate in the $2\theta$ range from 2° to 65°.
2. Semi-orientated aggregate heated at about 200°C at least for an hour, in the $2\theta$ range from 2° to 15°.
3. Ethylene glycol treated aggregate in the $2\theta$ range from 2° to 10°.

Pattern No. 3 of the above list was made only for confirmation of montmorillonite mineral, and pattern No. 2 was mainly used for discrimination of halloysite from illite. Some representative patterns (No. 1 and No. 2) of each kind of fireclay are shown on Figs. 22, 23 and 24.

These figures are very characteristic. For instance, as shown on Fig. 22, the upper four patterns of untreated "kaolin" show distinctive reflections of about 10Å and 7.3A with some difference of intensity, and usually accompany continuous diffusion band between the two reflections. But 10Å reflection and intermediate diffusion band disappeared by heating at about 200°C, and only a sharp reflection of about 7.3Å remained.

These facts suggest that 10Å peak is 001 reflection of halloysite and 7.3Å peak that of metahalloysite. It is probable that the intermediate diffusion is the result of mixing of partly dehydrated halloysite. But X-ray diffraction patterns of "kaolin" samples from other than Obora do not always show 10Å reflection, but 7.3Å reflection with asymmetrical diffusion at the lower angle

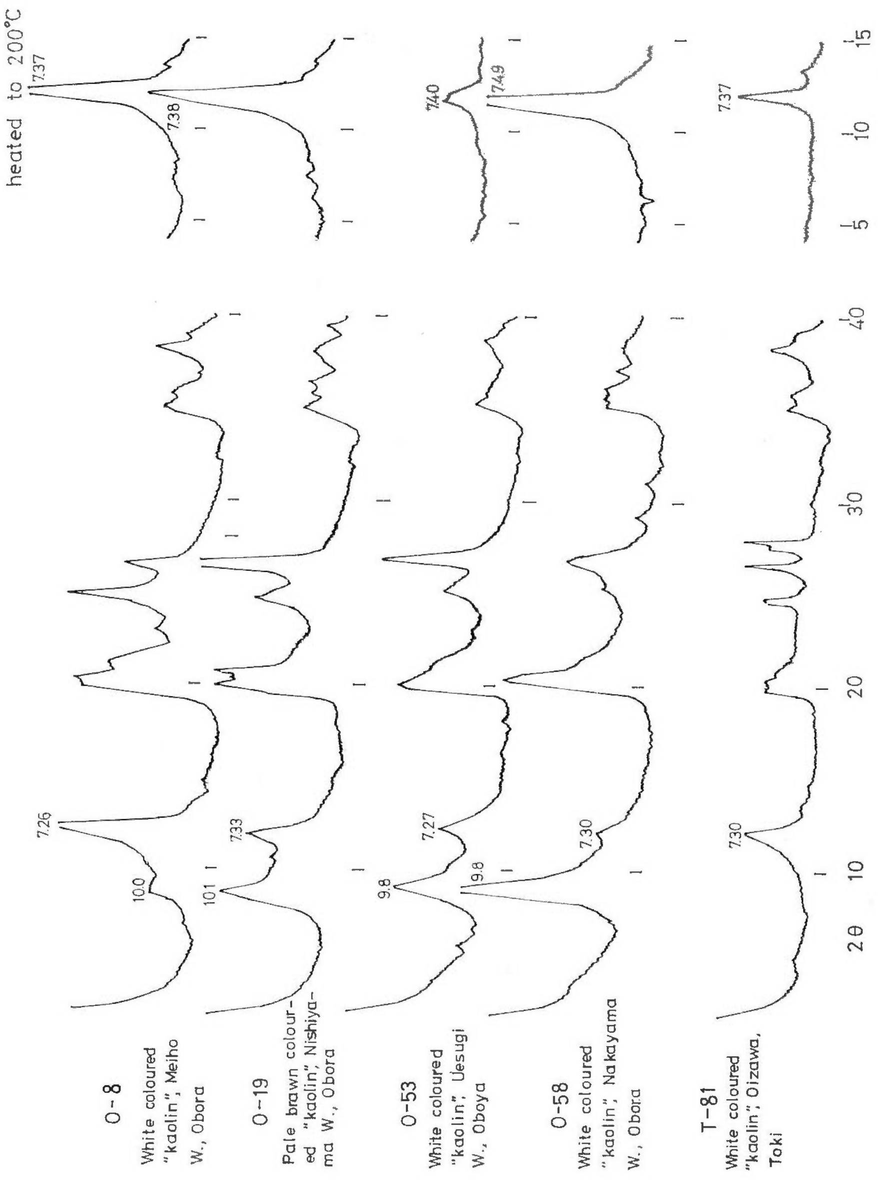



Fig. 22 X-ray powder patterns of the so-called kaolin

(Fig. 22: T-81). Since it is also destroyed by heating at about 200°C, this diffusion could be caused by mixing of halloysite dehydrated to some extent.

Next relating to gaerome-clay (clay fraction), there may be variable types of X-ray diffraction patterns. The one type of patterns show reflection at about 7.3Å with a remarkable diffusion at the lower angle, and another shows a sharp reflection at about 7.2Å with little diffusion. But generally speaking, a tendency that the lower the 7.2Å peak the broader the accompanying diffusion becomes is observed on Fig. 23. Since it disappeared after heating at about 200°C, the broad diffusion band at the lower angle of 7.3Å reflection is concluded to be from partly dehydrated halloysite (T-38, T-59). On the other hand, the X-ray patterns, the 7.2A reflection of which is stronger than the 4.45Å reflection, are from kaolinite. Because kaolinite, usually of a platy form, tends to be more preferrentially orientated than halloysite of lath or tubular form, and so its basal reflection of 7.2Å should be enhanced more strongly than the 020 reflection at about 4.45Å (Souza SANTOS etc., 1964). The upper three patterns on

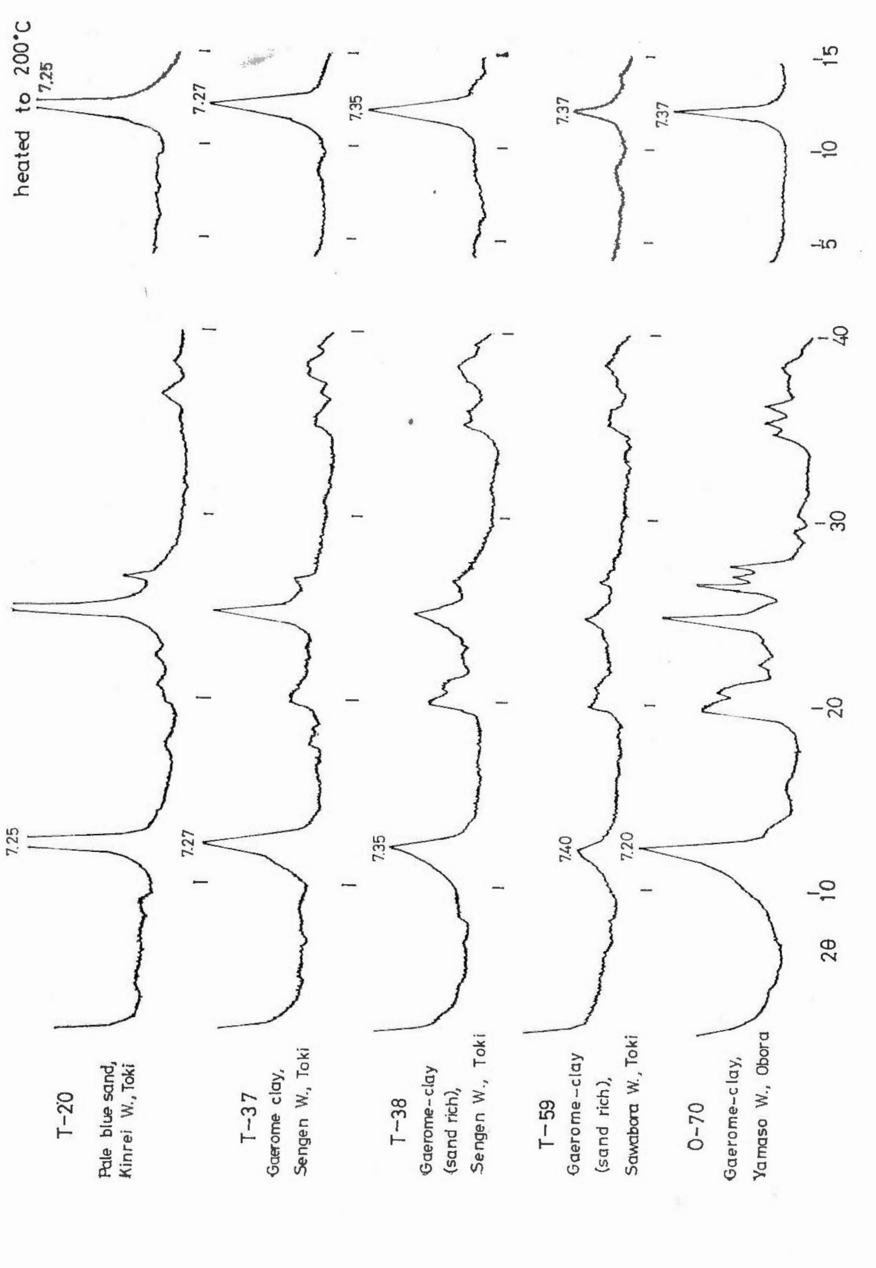



Fig. 23 X-ray powder patterns of gaerome-clay (washed)

Fig. 23 (T-20, 37 and 38) have not only similar characteristics as stated above, but also two groups of triplets in the $2\theta$ range from 35° to 40° though in a blurred appearance. These facts suggest that these samples contain more or less kaolinite, probably of the disordered type since their hkl reflections between 20° and 22° are not always distinctive.

In conclusion, it may be said that gaerome-clay (clay fraction) consists of mixture of kaolinite and halloysite in variable ratio.

Lastly, relating to kibushi-clay, it is very characteristic that most of the diffraction patterns show not only the reflections arising from kaolin minerals, but also those from the other minerals. As shown on Fig. 24, all powder patterns have the reflection at about 15Å. They can be identified with the 001 reflection of montmorillonite since their spacings were expanded to 16 or 17Å after solvation with ethylene glycol.

And some patterns show weak reflection bands at about 10Å. Since they did not disappear by heating at about 200°C, it may be assumed that they arose from a small amount of illite. Besides considerable amount of quartz are contained in all kibushi-clays. Most of kaolin minerals in kibushi-clay may be identified with disordered kaolinite from the striking powder patterns as follows.

1. Compared with 020 reflection at about 4.45Å, the 001 reflection at about 7.2Å tends to be enhanced remarkably.
2. Two groups of triplets in the $2\theta$ range from 35° to 40°, which are indicative of kaolinite, can be observed on some patterns (N-98, O-42).
3. A few reflection bands in the $2\theta$ range from 20° to 22°, which are indicative of well-ordered kaolinite, are not separated from each other.

But one pattern (N-84) does not show the 2nd indicative feature stated above. Therefore it is difficult to determine whether this sample (miscellaneous clay) consists mainly of kaolinite or metahalloysite.

### VI. 2. 2 Differential thermal analysis and thermogravimetric analysis

**Method**

DTA and TGA curves of each kind of fireclay samples, X-ray powder patterns of which are illustrated in Fig. 22, 23, and 24, are shown on Fig. 25, 26 and 27 correspondingly. These investigation were carried out by the Differential Thermal Balance made by Rigaku Denki Co., Ltd.

Experimental conditions were fixed as follows,

| | |
|---|---|
| Specimen weight | : 0.300 g |
| Thermocouples | : Pt-Pt.•Rh |
| Sensitivity of DTA | : $\pm 50\ \mu V$ |
| Sensitivity of TGA | : $\pm 50$ mg |
| Heating rate | : about 10°C/min. |
| Chart speed | : 180 mm/hr |
| Atmosphere | : air |
| Maximum temperature | : 1,180°C |

**Results and Discussions**

All DTA curves of "kaolin" appear to be similar to that of typical halloysite. And the differences in height of the endothermic peaks at about 120°C is due to the differences of dehydration grade of halloysite. The size of 120°C peaks

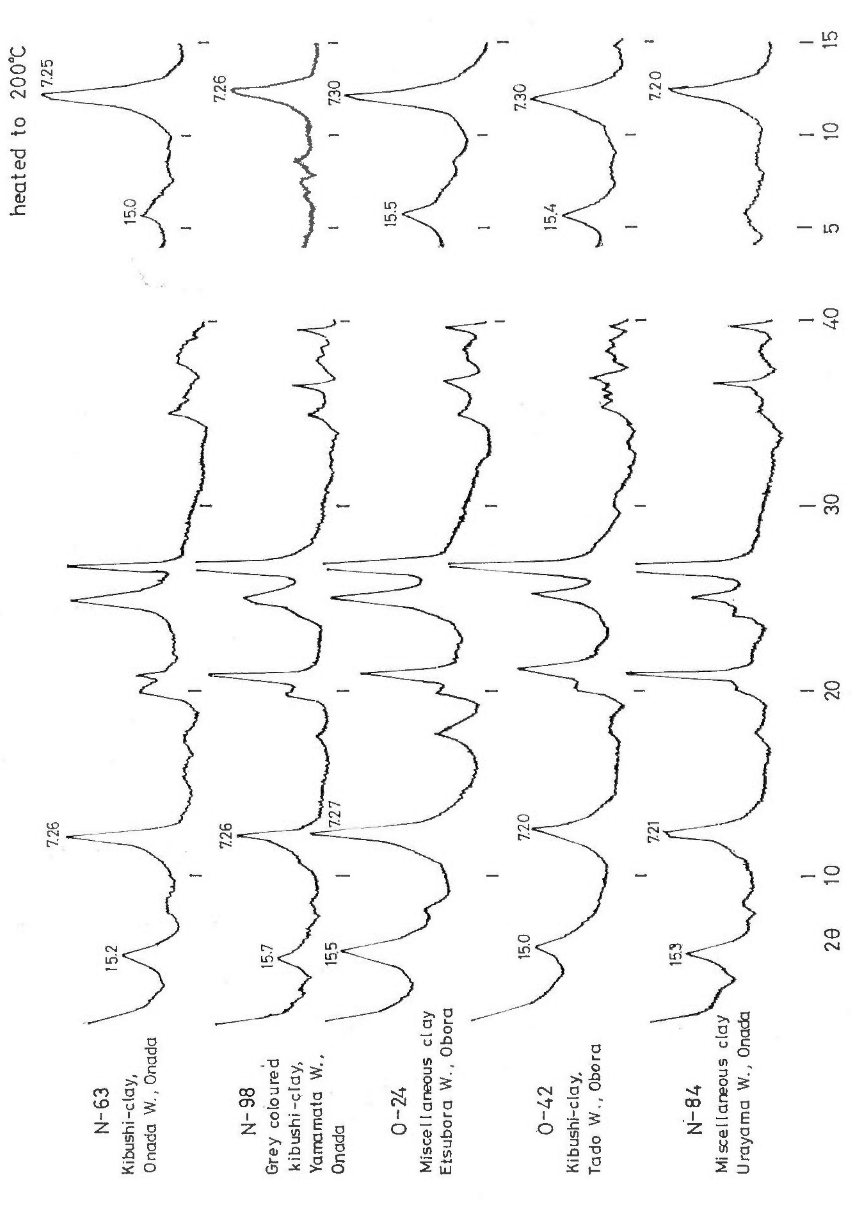



Fig. 24 X-ray powder patterns of kibushi-clays

of "kaolin" seems to be not only directly proportional to the weight loss at the temperature range from 50°C to 200°C, but also seems to reflect the height of 10A peak in its X-ray pattern (Figs. 22, 25).

As regards gaerome-clays, each DTA curve does not always correspond to the result of X-ray analysis. For example, DTA curve of sample T-38, which appears to contain more halloysite than T-20 and T-37 from results of X-ray analysis, has only a small endothermic peak at about 120°C.

DTA curves of kibushi-clays have some distinct characteristics, which are

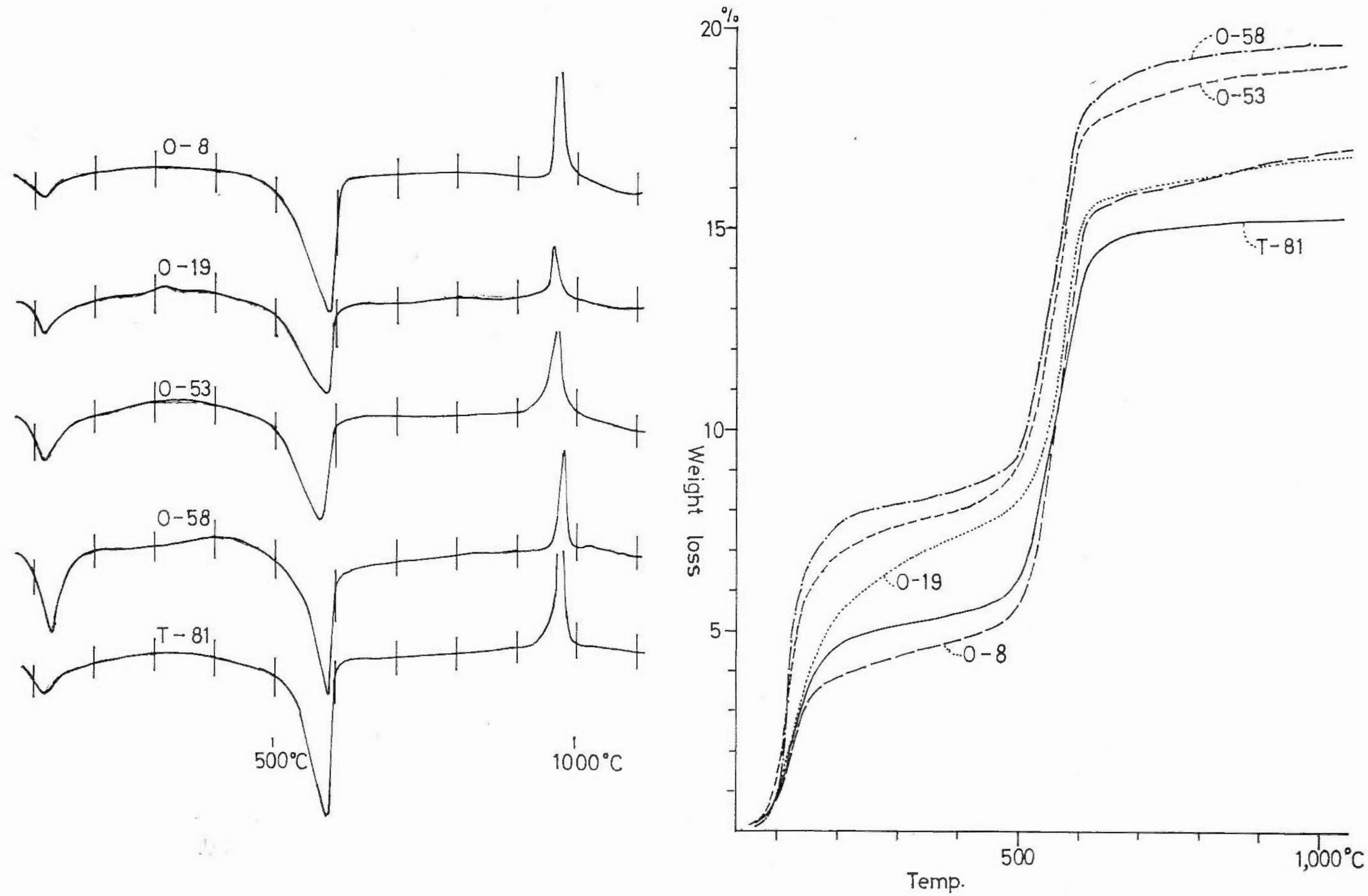



Fig. 25 DTA and TGA curves of the so-called kaolin

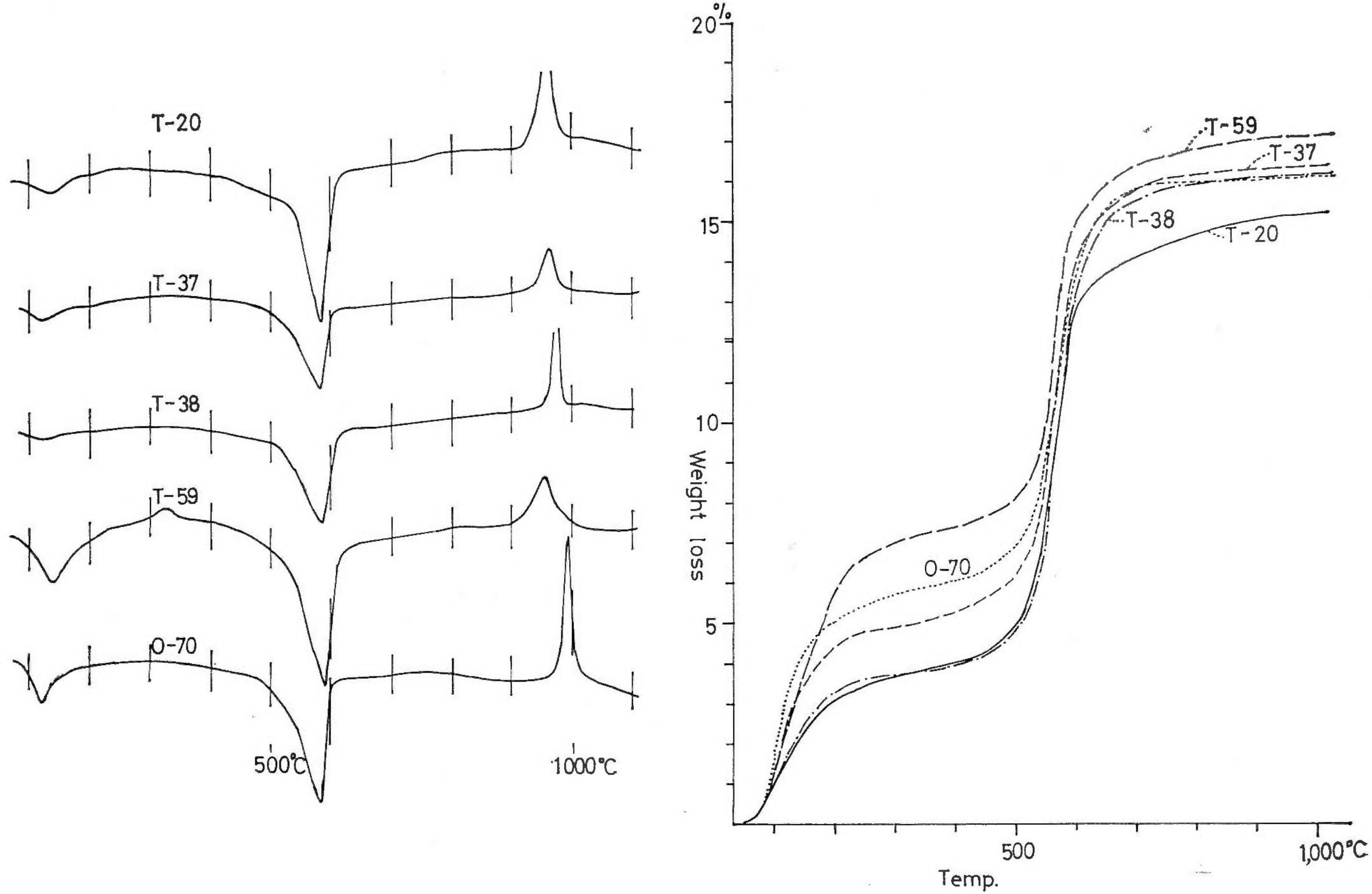



Fig. 26 DTA and TGA curves of washed gaerome-clays

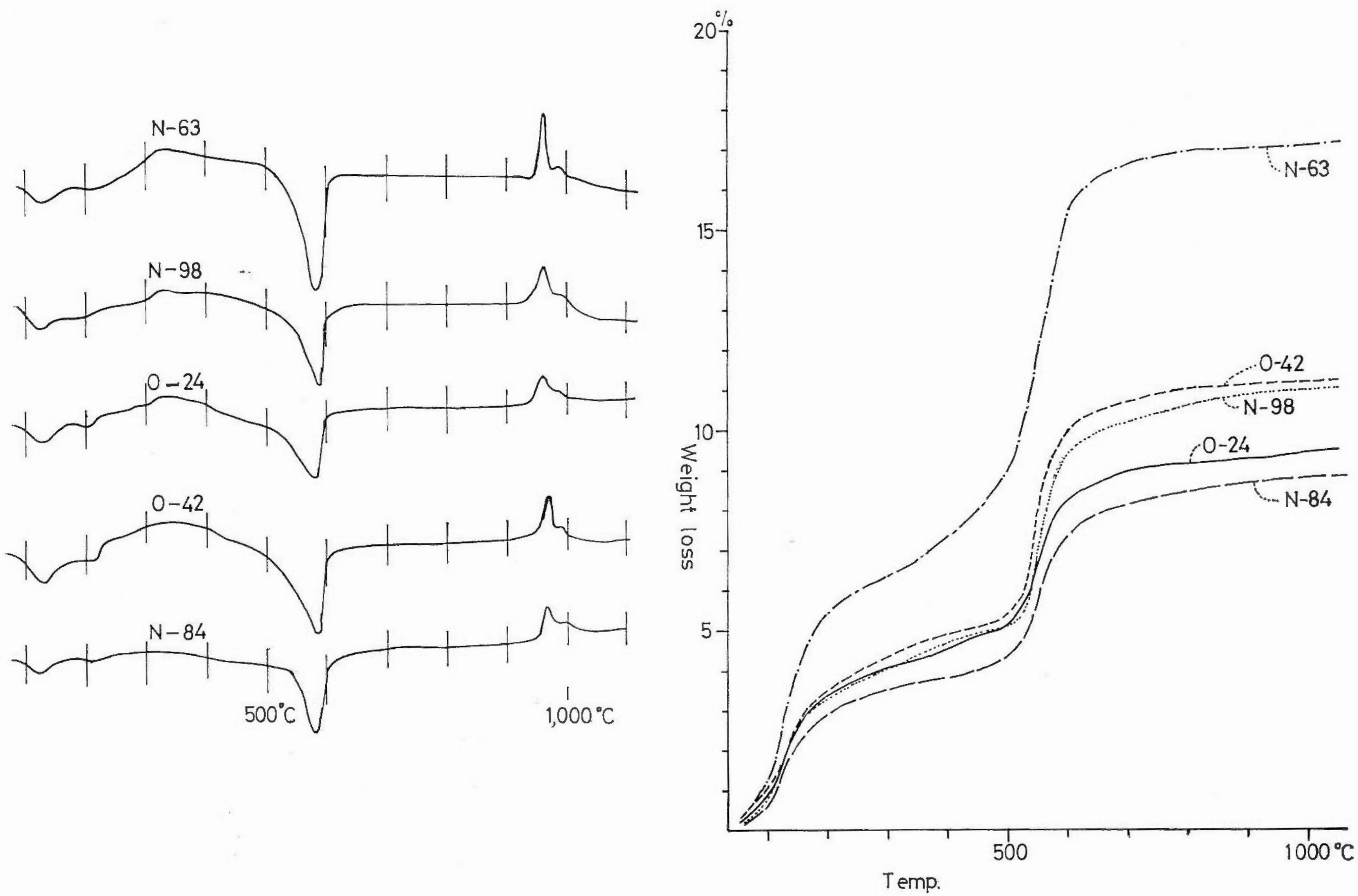



Fig. 27 DTA and TGA curves of kibushi-clays

never observed in those of the other fireclays. For example, both a small endothermic peak of about 200°C, adjacent to the peak of about 120°C and a small exothermic peak of about 990°C, accompanied by the peak at about 960°C, are believed to be the effects caused by thermal reaction of montmorillonite. Other notable characters are that most of the exothermic peaks in the range from 950°C to 990°C are smaller in size and appear at lower temperatures (960~955°C) than those (970~980°C), of halloysite as illustrated in Fig. 25. Such exothermic reaction may not only be cause by mixing of impure materials, but also by individual characteristics of the kaolin mineral contained in kibushi-clay. For example, since one sample (N-63) containes little quartz and consists dominantly of disordered kaolinite, its DTA curve should represent the character of kaolinite.

The relationship between the exothermic reaction of kaolin minerals and their crystalinity will be considered in later way. Therefore it would not be presented here, beyond the description of results of thermal experiments for each fireclay.

At all events, it can be said that DTA curves of each fireclay vary greatly and reflect its mineral composition as a whole.

### VI. 2. 3 Electron microscopic observation

As stated above, kibushi-clay, gaerome-clay and "kaolin" all showed different characteristic in both X-ray and thermal analysis. Electron microscopic observation also showed that each of them appears in different form of crystals.

Most of "kaolin" samples generally contain lath form crystals, which consist of parallel aggregate of flaky crystals about 0.2~0.5 microns in length. Exceptionally, a few platy crystals were observed in electron micrograph (Plate IV-2). Moreover, electron micrographs of the sample O-19, occurring with lignite, and the sample T-81, which does not show 10Å reflection in its X-ray pattern, are also similar to that of common "kaolin" (Plate V-1, 2). These facts may show that "kaolin" is largely composed of mixture of halloysite and metahalloysite.

Relating to gaerome-clay, crystal forms of kaolin minerals vary with different samples. For instance, an electron micrograph of the sample T-59, X-ray pattern of which shows 7.3Å reflection with broad diffusion at the region of lower angles, showed that it consisted mainly of tube-like crystals of halloysite about 0.5 microns in length (Plate VI-1). On the other hand, electron micrographs of the sample T-20 and T-37, which probably contain abundant kaolinite in view of their X-ray diffraction patterns, showed that they consisted largely of platy crystals with irregular outlines about 0.1 microns in diamiter (Plate VI-2, VII-1). Above facts show that some gaerome-clays contain abundant kaolinite and others halloysite. Ratio of kaolinite to halloysite in gaerome-clay seems to differ with different samples.

But neither of platy kaolinite and tube-like halloysite seem to be contained very much in a gaerome-clay sample, O-70, taken at the northern part of Obora sub-basin. It consits dominatly of globular crystals of halloysite about 0.3 microns in diameter (Plate VII-2). It is said generally that globular particles of halloysite are of poor crystalinity and most of them may have been originated from glassy materials. Therefore, above fact may show that gaerome-clay occurring at Obora has been mixed with some volcanic glasses.

In electron micrographs of kibushi-clay, all kaolin minerals are not of tube-like form but of platy form with irregular outlines 0.1 to 0.2 microns in diameter (Plate VIII-1, 2). But a miscellaneous clay, the sample N-84, appears to contain small amount of tube-like halloysite.

The result of observation stated above seem to agree with the inference based on X-ray and thermal experiments.

### VI. 3 Mineral composition of fireclays

All clay samples taken in this district were investigated mainly by X-ray diffraction analysis. Identification of each kind of kaolin minerals were carried out according to the following criteria;

1. The existence of heating change of the 10Å reflection and diffusing band near the 7.2Å reflection.
2. The existance of preferential orientation giving enhancement of basal reflections.
3. The presence of two groups of triple peaks in the $2\theta$ range from 35° to 40°.

And mineral compositions of clay samples deduced mainly by X-ray analysis were investigated quantitatively by means of rapid chemical analysis of $SiO_2$, $Al_2O_3$ and Ig. loss. These investigations gave a conclusion with respect to mineral composition of each fireclay as follows.

(1) Kibushi-clay is composed mainly of disordered kaolinite and fine-grained quartz, frequently accompanying montmorillonite and small amount of illite.

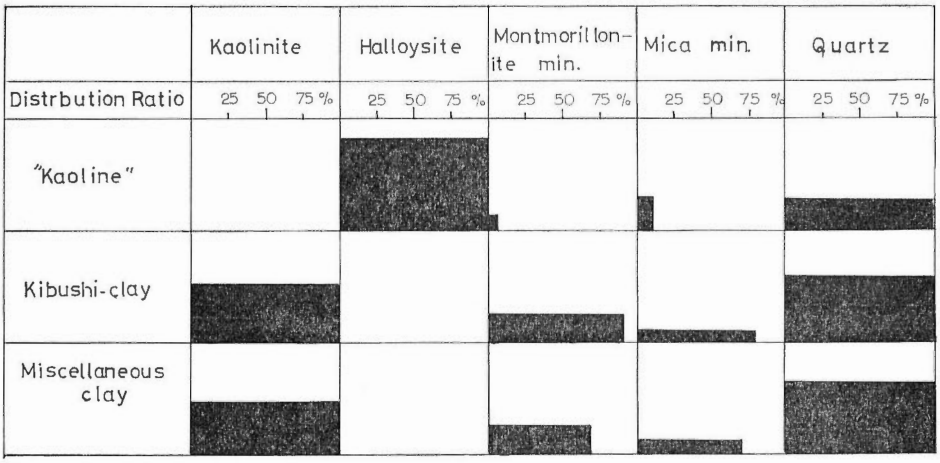
(2) The so-called kaolin consists dominantly of mixture of halloysite and metahalloysite with a little quartz and volcanic glass.

It is noteworthy that the two kinds of fireclay, mineral composition of which differ with each other, are intercalated in the similar formation. Fig. 28 was made in order to illustrate the mineral compositions of a few types of fireclays occurring at Obora area, which are the so-called kaolin, kibushi-clay and miscellaneous clay. In Fig. 28, the height and width of the black rectangles mean the average content and distribution frequency respectively of a component mineral in a fireclay. And this figure shows that mineral compositions of kibushi-clays are nearly similar with those of miscellaneous clays but differ distinctly with those of "kaolin". These facts would prove that "kaolin" was derived from different source materials from those of the other clays.

Making an additional remark, kibushi-clay generally contains more montmorillonite than miscellaneous clay. In other words, montmorillonite tend to be concentrated in kibushi-clay.

(3) Next, gaerome-clays consists mainly of kaolin mineral and quartz with small amount of feldspar and mica clay minerals. But some of them contain abundant halloysite and the other kaolinite, and the ratio of halloysite to kaolinite in gaerome-clay is not constant.

Generally, relating to gaerome-clay and pale blue fine sand occurring in the lower horizon at Toki basin, it can be said that the samples including abundant sand fraction contain more halloysite, and while the samples with large clay fraction tend to include more kaolinite. The tendency stated above is illustrated in Fig. 29. This figure shows the relationship between the clay fraction percentages of gaerome-clay and pale blue fine-sand and the shape of the




Remarks: height of painted rectangle shows mean percentage of each mineral roughly estimated by x-ray powder patterns

Fig. 28 Mineral distribution in each type of fireclay at Obora area

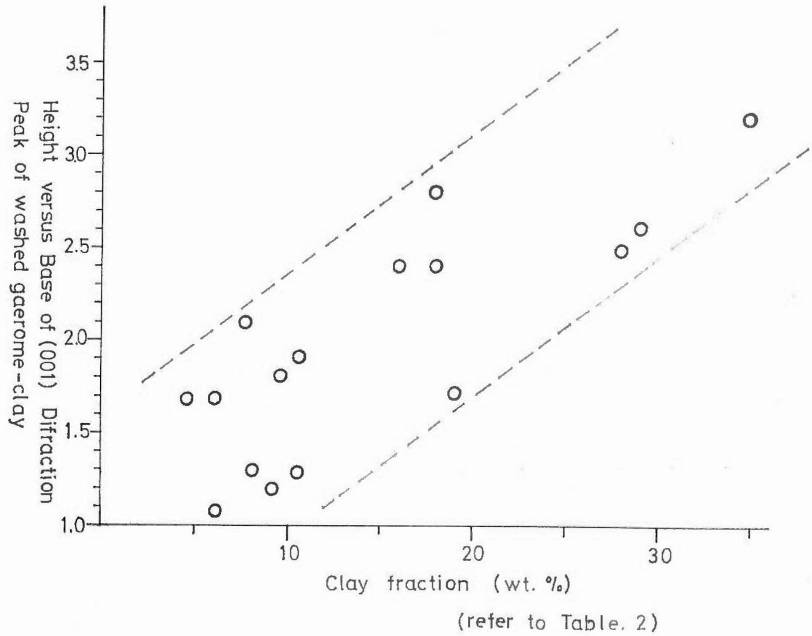



Fig. 29 Relationship between the shapes of the 001 diffraction peaks and the clay fraction percentages in gaerome-clay, Toki area

7.2Å peak in their X-ray diffraction patterns. The clay fraction percentages were roughly measured by washing test. And the shape of the 7.2Å peak was represented by the ratio of height (Ih) versus base (Iw) of this reflection on their X-ray patterns. Because the more the sample contains halloysite the broader the diffusion band at the lower angle of the 7.2Å peak, and on the other hand the more the clay includes kaolinite the stronger the basal reflections. Therefore the ratio of height versus base of the 7.2Å peak (Ih/Iw) may generally reflect the ratio of kaolinite and halloysite in the sample.

Fig. 29 shows the tendency that the more the sample contains clay fraction the larger its Ih/Iw value becomes. In other words, it can be said that better-sorted gaerome-clays include more kaolinite than ill-sorted ones, and the latter usually contain more halloysite. As shown by electron micrograph, many kaolinite particles are of about 0.1 microns in diameter and many halloysite crystals are about 0.5 microns in length. It may be reasonably surmised that the difference of their particle size caused their segregation throughout transportation and deposition.

### VI. 4 Chemical composition and refractoriness

In order to know some natures for application, rapid chemical analysis and measurement of refractoriness were carried out for most of specimen used in this study. Results of these investigation will be described in detail on another paper, and their general description will be stated here.

The so-called kaolin has the most constant nature among the fireclays in this district. Especially, "kaolin" clay taken at Obora area has little impurities, and is mostly of SK 34 to 36 in refractoriness. But most of "kaolin" taken elsewhere contain some impurities and are of lower refractoriness.

Being separated usually by washing, the properties of gaerome-clay are relatively constant. Their refractoriness seem to be higher than SK 33 in most cases.

Lastly relating to kibushi-clays, a few have SK 34 in refractoriness, but most of them are not so high. It may be considered that their refractoriness are influenced by impure minerals, which are quartz, montmorillonite and illite. Especially, illite seems to cause remarkable drop of refractoriness. Many miscellaneous clays also appear to be similar in chemical composition and refractoriness to inferior kibushi-clay.

### VI. 5 Summary

It is very notable in many investigation described previously, that "kaolin", gaerome-clay and kibushi-clay each has its own characteristics concerning fabric, mineral composition, type of main kaolin mineral, refractoriness and so on. Their characteristics may be summarized as follows.

#### VI. 5. 1 "Kaolin"

The so-called kaolin clays are generally micro-grained and homogeneous, and some of them retain the texture of tuff. They consist dominantly of mixture of halloysite and metahalloysite with small amount of quartz and volcanic glass. Halloysite crystals appear mostly to be parallel aggregates of rectangular laths from 0.1 to 0.3 microns in length. Most "kaolin" are more than SK 33 in refractoriness.

### VI. 5. 2 Gaerome-clay

It is believed that gaerome-clays were mostly derived from weathered and decomposed granitic rocks. They are composed mainly of kaolin minerals and quartz grains with small amount of feldspar and mica. Clay fraction percentages in gaerome-clays are very variable (5 to 30%). Moreover, some of gaerome-clays contain large amount of kaolinite as main clay component, and others include more halloysite. But concerning gaerome-clays taken at Toki area, it can be said that the clay fraction percentage generally reflect the ratio of kaolinite versus halloysite.

Kaolinite particles, most probably of the disordered type, are six-sided plates with irregular outlines and about 0.1 microns in diameter. Most halloysite particles appear to be tube-like or rectangular in shape of about 0.5 microns in length. But as stated previously, globular halloysite particles were also observed in an electron micrograph of a gaerome-clay sampled at the northern part of Obora area. Generally gaerome-clay separated by washing is more than SK 33 in refractoriness.

### VI. 5. 3 Kibushi-clay

Kibushi-clays are generally blackish brown to dark brown and very plastic. Microscopic observation shows that many kibushi-clays are characterized by dendritic, mesh or fibrous fabrics.

Most of them consist mainly of disordered kaolinite and fine-grained quartz accompanying montmorillonite and small amount of illite. Kaolinite particles seem to be of six-sided platy form with irregular outline 0.1 to 0.3 microns in diameter. Mineral compositions of miscellaneous clays are also generally similar to those of kibushi-clays. But it is likely that montmorillonite tends to be more concentrated in kibushi-clay than in miscellaneous clay.

These clays are SK 26 to 34 in refractoriness, but a few are more than SK 31.

## VII. Origin of Fireclays

### VII. 1 The relationship of the mineral distribution in the Tokiguchi formation to its sedimentary process

In order to know the genesis of the fireclay deposits, it is necessary above all to understand the relationship between the geological occurences of fireclays and their mineral compositions. Since all fireclay deposits occures in the Tokiguchi formation, the sedimentary process of this formation must have close relation with formation of the fireclay beds.

Fig. 30 illustrates the distribution of clay minerals corresponding to the stratigraphical sequences of the Tokiguchi formation in each basin. As shown in Fig. 30, gaerome-clay facies were deposited at the opening period of deposition of the Tokiguchi formation. These facies generally contain kaolinite and partially dehydrated halloysite as main clay components, but include little mont-

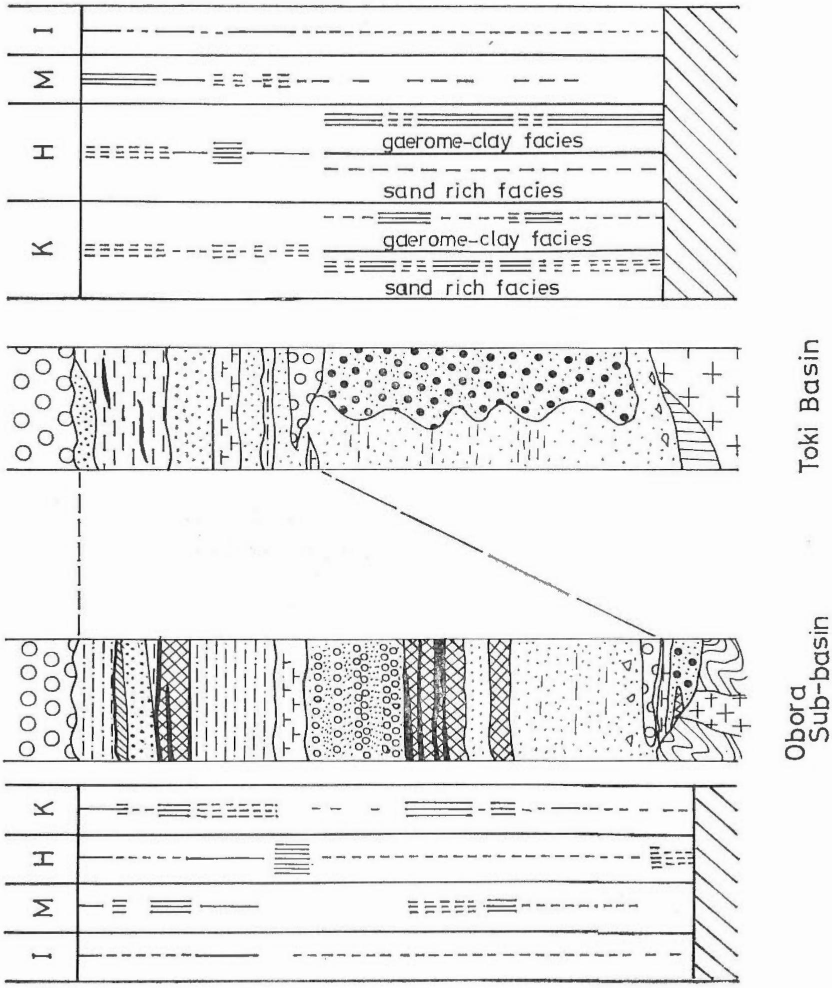



Fig. 30 Relationship between the clay minerals distribution and the stratigraphical sequences of the Tokiguchi formation

morillonite.

Such mineral distribution appears to have gone through notable change after the deposition of the chert gravel beds.

At Obora sub-basin, the clay rich facies and the sand and granule rich facies, overlaping the chert gravel bed, usually contain kaolinite and montmorillonite. But halloysite seems to be distributed only in tuff beds including the so-called kaolin.

Similarly at Toki basin, the upper clay and sand rich facies generally include montmorillonite and kaolin minerals. But kaolinite and halloysite seem to be distributed together.

Accordingly, the chert gravel beds may be very significant for the purpose of understanding the geological meaning of clay minerals distribution in the Tokiguchi formation. That is, depositional area of this formation was rapidly enlarged as soon as the chert gravel beds were deposited, and since that time volcanic materials began to be supplied into some basins. On the other hand, the upper facies underlain by the chert gravel beds contain relatively abundant montmorillonite, and intercalate a few halloysitized tuff beds.

It appears as if these facts suggest that montmorillonite was originated from volcanic ash. Because it is a generally accepted theory that volcanic materials deposited in marine environment tend to be montmorillonitized diagenetically (WEAVER: 1958, SUDO: 1954, MUKAIYAMA etc.: 1964).

But it was shown previously in Fig. 28 that montmorillonite tends to be more concentrated in kibushi-clay than in miscellaneous clay. Kibushi-clay, which is accompanied by lignite seam, should have been aged under weakly acidic environment influenced by organic acid originating from lignite. Therefore there are few possibilities that montmorillonite was formed authigeneously. It is reasonably considered that montmorillonite minerals were transported as detrital materials and deposited coagulating rapidly in acidic environment.

It has been well known that montmorillonite is relatively stable during surface weathering and sometimes occures as a secondary sediment (HARRISON and MURRAY: 1959, ROBERTSON: 1961).

As stated often, in this district authigenic minerals derived from volcanic materials may be mostly halloysite, which is the main component of "kaolin".

It has been reported frequently that volcanic glasses tend to be halloysitized by weathering (SUDO and OSSAKA: 1952, MUCHI: 1962 and others). Thus it is expected that they can be halloysitized similarly under fluvio-lacustrine environment. Similar assumption was presented previously by KITAZAKI and ARAKI (1952), AKAMINE (1954), NOZAWA (1955b) and others.

But such assumption does not always signify that all halloysite minerals in the Tokiguchi formation were derived from volcanic materials. Because some gaerome-clay, which contain little volcanic materials, include a large amount of halloysite, too. In order to understand the genesis of these kaolin minerals, it is necessary to know the conditions which prevailed on the basement rocks at the time of deposition of the Tokiguchi formation.

## VII. 2 Weathering effects preserved in the basement rocks

### VII. 2. 1 General features

In Tajimi-Toki district, it is frequently observed that the basement rocks were partially subjected to remarkable alteration. And the results of careful survey show that these altered zones seem to be restricted to the basement rocks overlain by the Pliocene sediments.

Especially the basement rocks covered directly by the Tokiguchi formation have been subjected to vigorous alteration without exception. And it is always observed that the upper part of basement rocks have been most remarkably altered and the altered rocks grade down into weakly alterd ones.

It is less possible that hydrothermal alteration occurred uniformly over such extensive region. Moreover such a geological evidence that these rocks were hydrothermally altered has not yet been found.

It is reasonably considered that these altered rocks were formed during weathering before the deposition of the Tokiguchi formation. Because a regional factor such as climate would be the best way of explaining similar occurrences of altered rocks in such wide region.

Relating to both the Palaeozoic and granitic rocks of the basement, weathering effects preserved in them can be observed precisely in some places. These observations will be presented later. But concerning the Miocene beds, it is difficult to distinguish the clay minerals formed in them during weathering with the ones developed authigeneously. Rough investigation showed that montmorillonite, metahalloysite, illite and randomly mixed layer mineral are contained in some Miocene rocks. Especially it has been reported that the Oidawara formation, the uppermost member of the Miocene group, contains abundant montmorillonite (NOZAWA: 1955a).

### VII. 2. 2 Occurrence of Onada silica-stone

At Onada of Tajimi area, white colored silica-stones are mined at the two workings, Maruyo and Okumura 1st. These silica-stones are very brittle. But it is observed that platy laminae peculiar to chert are preserved in them. And at Maruyo workings, it can be observed that silica-stone grades downward into weakly altered chert stained by iron hydroxide (Fig. 31). These occurrences show that silica-stone was derived from chert. The altered chert workable as silica-stone is usually about 5 meters thick, and always occurs at the uppermost part of the Palaeozoic rocks covered directly by the Tokiguchi formation. And this formation contains not only breccias of white silica-stone, but also those of unaltered chert. These facts suggest that white silica-stones had been formed at the surface of the basement before the Tokiguchi formation was deposited.

Microscopic observation also shows that the silica-stone has similar cryptocrystaline texture with that of chert.

But comparing the chemical compositions of a silica-stone and an unaltered chert, it is detected that alteration caused a considerable decrease in $SiO_2$ and $Fe_2O_3$ and a little increase in $Al_2O_3$ and $H_2O$ (Table 3). Similar silica-stones with

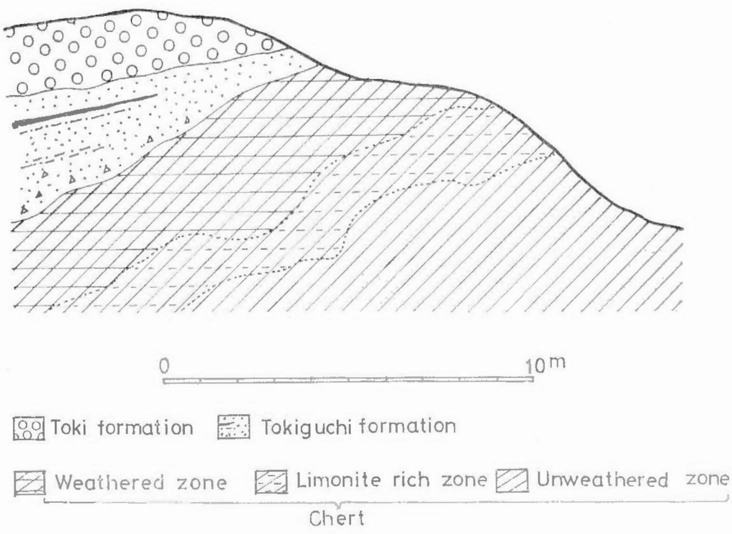



Fig. 31 Occurrence of the silica-stone deposit at Maruyo mine, Onada

those of Onada area are mined at a few other workings, which are Toyane and Ishihiro at the southern part of Tajimi-shi.

Table 3 Chemical composition of the original chert and the weathered one at Maruyo workings, Onada

| | Original chert (wt. %) | Weathered chert (wt. %) |
|---|---|---|
| $SiO_2$ | 97.52 | 93.86 |
| $TiO_2$ | 0.05 | 0.12 |
| $Al_2O_3$ | 0.67 | 3.37 |
| $Fe_2O_3$ | 0.38 | 0.17 |
| FeO | 0.16 | 0.16 |
| MgO | 0.10 | 0.18 |
| CaO | 0.01 | 0.01 |
| $Na_2O$ | 0.01 | 0.03 |
| $K_2O$ | 0.02 | 0.05 |
| $+H_2O$ | 0.37 | 1.28 |
| $-H_2O$ | 0.16 | 0.22 |
| Total | 99.45 | 99.45 |

Analyst : Masaki KAWANO

## VII. 2. 3 Weathered granite at Kakino mine

### Geological occurrence

Kaolin clay, probably formed in granitic rocks by weathering, is worked at Kakino mine which is situated at the southern end of Toki-shi.

Since it is used as raw material for pottery and white ware, this clay is called Kakino "kaolin" (FUKUO and KUCHINA : 1960). But it is residual kaolin and differs distinctly in genesis with "kaolin" occurring at Obora area. Similar kaolin deposits are distributed at some places in this region.

The Seto group is distributed in the vicinity of Kakino mine, underlain unconformably by granitic rocks. This group consists dominantly of thick gravel bed with a thin bed of gaerome-clay and pale blue sand. The gravel bed may be correlated with the Toki formation, and the gaerome-clay bed with the Tokiguchi one.

Most of the granitic rocks are leucocratic and coarse-grained biotite granite intruded by dioritic dikes at some places. And in some localities the xenoliths rich in biotite are observed in granite. Granitic rocks seem to be generally affected by weathering and made brittle to considerable depth. But kaolinized granite appears to occur mainly at the places, on which the gaerome-clay and sand beds overlie (Fig. 32). The kaolinized granite is illustrated in the profile of Kakino mine (Fig. 33). As shown in this figure, well-kaolinized zone, which is restricted at the uppermost part of a few meters thick, grades downward into weakly weathered zone through intermediate one stained by iron hydroxide.

But the gravel bed on the kaolinized granite also contains some argillized gravels of quartz prophyry. This fact shows that weathering effects after deposition of the gravel bed are not negligible (FUKUO and KUCHINA: 1960).

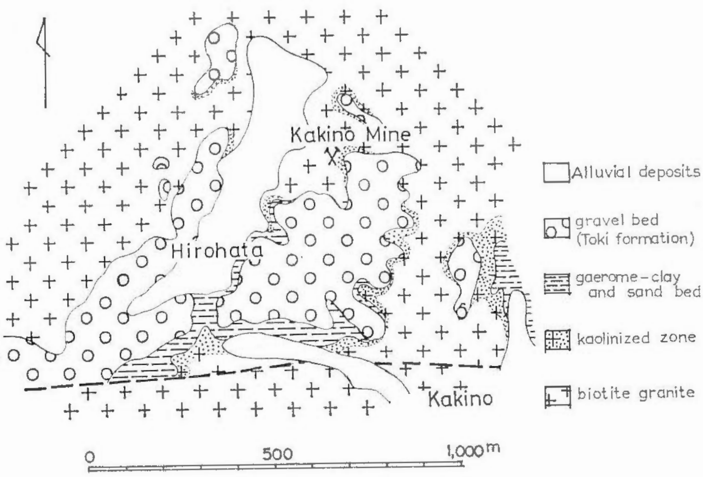



Fig. 32 Geological map of Kakino area

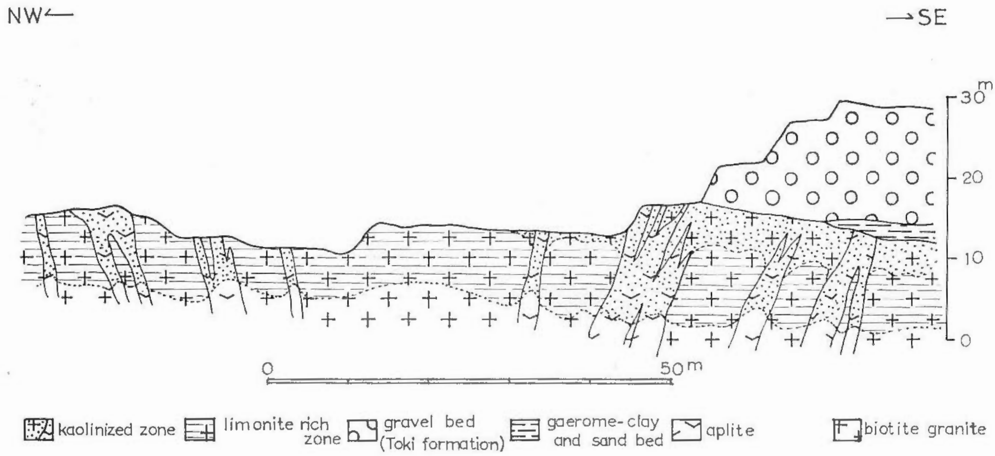



Fig. 33 Geological profile of residual kaolin deposit at Kakino mine

Nevertheless, it is reasonably considered that the kaolinized granite was mostly formed by weathering before the deposition of the gaerome-clay and sand beds, because the top parts of granite covered by the gravel bed are not always kaolinited.

### Weathering effects

In order to know the detailed weathering effects on granite, the differences in the mineral and chemical components of the original and kaolinized granite were investigated by microscopic and electron microscopic observations, X-ray diffraction and chemical analysis.

Microscopic observation of weathered granite showed that plagioclase and biotite were most remarkably altered. Especially, most of plagioclase were altered into aggregates composed mainly of clay minerals. Many biotite appears to have been divided into some flakes elongated in the direction perpendicular to the cleavage. And a few flakes have the so-called vermicular form. Most of them have been altered into biotite pseudomorphs, probably consisting mainly of kaolin mineral, which shows little pleochroism.

Next, many potash feldspars appear to be minutely traversed by mesh-like cracks, but retain visible microcline texture. Lastly quartz grains do not seem to have been subjected to alteration at all (Plate IX-1).

Such observations are in accord with the results of chemical analysis, which are laid out in Table 4. Table 5 indicates quantitative increase and decrease of each component caused by weathering of granite. As shown in the two tables, alkali elements are nearly constant in quantity. But most of CaO and MgO were leached throughout weathering process. Total iron also decreased, and about 30 percent of $SiO_2$ seems to have been leached.

On the other hand, two components of $Al_2O_3$ and $H_2O$ increased remarkably during weathering process.

It can be easily assumed that decomposition of biotite and Ca-rich plagioclase gave rise to such decrease of some components. But increase of $Al_2O_3$ was likely caused by removal of aluminium hydroxide, probably originating from altered Ca-rich plagioclase.

Next, X-ray analysis and electron microscopic observation showed that weathered granite contains both platy kaolinite and tube-like halloysite. Moreover, it was ascertained that biotite rich granite contains much kaolinite as weathering products, compared with the leucocratic one (Plate IX-2, X-1, 2). The fact that both kaolinite and halloysite coexist in the weathered granite would be very useful to understand the variable kaolin mineral composition in gaerome-clays presented previously.

Table 4 Chemical composition of the original granite and the kaolinized one at Kakino mine

| | Original granite (wt. %) | Kaolinized granite (wt. %) |
|---|---|---|
| $SiO_2$ | 76.74 | 60.86 |
| $TiO_2$ | 0.17 | 0.23 |
| $Al_2O_3$ | 12.01 | 23.18 |
| $Fe_2O_3$ | 0.48 | 0.83 |
| FeO | 1.22 | 0.04 |
| MgO | 0.38 | 0.12 |
| CaO | 1.63 | 0.01 |
| $Na_2O$ | 0.52 | 0.50 |
| $K_2O$ | 6.00 | 6.84 |
| $P_2O_3$ | —— | —— |
| $+H_2O$ | 0.44 | 5.63 |
| $-H_2O$ | 0.20 | 1.24 |
| Total | 99.79 | 99.48 |

Analyst : Masaki KAWANO

Table 5 Isovolumetric calculation

| | Amount in 1 cm | | Absolute difference (mg) |
|---|---|---|---|
| | Original granite (mg) | Kaolinized granite (mg) | |
| $SiO_2$ | 1,980 | 1,455 | −525 |
| $TiO_2$ | 4 | 6 | neg. |
| $Al_2O_3$ | 310 | 554 | +244 |
| $Fe_2O_3$ | 12 | 20 | +8 |
| FeO | 31 | 1 | −30 |
| MgO | 10 | 3 | −7 |
| CaO | 42 | neg. | −42 |
| $Na_2O$ | 13 | 12 | neg. |
| $K_2O$ | 155 | 163 | neg. |
| $+H_2O$ | 11 | 135 | +124 |
| $-H_2O$ | 5 | 30 | +25 |
| Weight of 1 $cm^3$ in milligram | 2,580 | 2,390 | −190 |

### VII. 2. 4 Agency of weathering

The weathering effects preserved in the basement rocks suggest that warm and moist climate prevailed on this region at the earlier Pliocene age. Similar assumption was already presented by MIKI (1949).

And lastly TOKUNAGA (1965) found some flora, which suggest sub-tropical climate, from the Tokiguchi formation at the southeastern part of Toki basin.

It can be said that a regional factor such as sub-tropical climate is the best way of explaining vigorous alteration of the basement rocks in such wide region.

### VII. 3 Discussion on the origin of Kaolin minerals

It has been presented at the first section of this paper that there are several different theories on the origin of kaolin minerals constituting kibushi-clays. They may be roughly divided into the two groups. One is the theory that considerable part of kaolin minerals have been formed authigeneously from volcanic materials (NOZAWA: 1955b, etc), and the other is the view that they are mostly detrital in origin (MATSUZAWA and others: 1960, etc).

It is surely considered from many evidences described previously that "kaolin" was derived from volcanic materials. But it is rather reasonably inferred that most of kaolin minerals contained in the other fireclays than "kaolin" are of detrital origin.

For example, relatively well-sorted gaerome-clay includes abundant kaolinite as main clay component, but many of ill-sorted ones are usually rich in halloysite. NOZAWA (1955b) presented the view that many kaolin minerals of tuff origin are halloysite and those derived from weathered granite are kaolinite. But it was detected in the present study that both kaolinite and halloysite coexist in remarkably weathered granite as Kakino "kaolin". Therefore it is considered that halloysite in gaerome-clay also has been derived from weathered granite. Many geological evidences show that active volcanism took place after deposition of the most part of gaerome-clay facies.

Relating to kibushi-clay, it is assumed by its mesh or dendritic fabrics that kaolin minerals have grown to some extent after their deposition. But most of them are of similar disordered type as those in miscellaneous clay which has no such characterisic fabrics. And so it is probable that basic lattice of kaolinite was formed before its deposition.

KITAZAKI and ARAKI (1952), NOZAWA (1955b) and others insisted that kaolin minerals in kibushi-clay were mostly derived from volcanic glasses, as well as those in "kaolin". But it is reasonably considered that the distinctive differences in mineral composition between the two mainly depend on the difference of source materials between the two. Therefore kaolin minerals in kibushi-clay may be mainly detrital in origin.

As stated previously, an abundance of kaolin minerals would have been formed in weathered granite of this region. It is most logically inferred that these kaolin minerals were concentrated in kibushi-clay. Moreover the Miocene rocks should have also played an important role as starting materials of kibushi-clay. Some of these rocks contain montmorillonite, kaolin minerals, illite and mixed-layer mineral as clay components. It is possible that these clay minerals also may have been mixed into kibushi-clays. Kibushi-clays of this district seem to contain relatively larger amount of impure minerals, compared with those of Seto district. (MATSUZAWA and others: 1960) It can be considered that such differences in mineral composition were mainly caused by the differences in basement rocks of the two districts.

## VIII. Conclusions

According to the geological and mineralogical studies stated above, the writer concludes on the genesis of the fireclay deposits in this district as follows.

(1) Some of the fireclays are composed mainly of authigenic kaolin minerals, and others are detrital in origin. Of the fireclays occurring in this district, "kaolin" is of the former type and gaerome- and kibushi-clays probably belong to the latter type.

(2) The so-called kaolin consists largely of mixtures of halloysite and metahalloysite with small amount of quartz and volcanic glass. It can be considered that it has been authigeneously derived from acidic tuff.

(3) Gaerome-clay is composed mainly of kaolin mineral and quartz grains accompanied by feldspar and illite. Its clay fraction percentage is very variable. And the gaerome-clays with large clay fraction contain more kaolinite, and ones with less clay fraction tend to include more halloysite. Such variation in mineral composition of gaerome-clays may have been caused by differential sedimentation owing to the difference in particle size of the two. Because kaolinite crystals are of platy form about 0.1 microns in diameter, and on the other hand most of halloysite crystals are of tube-like or rectangular form about 0.5 microns in length. And halloysite particles in "kaolin" are mostly parallel aggregates of lath crystals and differs in shape with those in gaerome-clay.

But a gaerome-clay sample taken at Obora area is an exception, and it contains globular form of halloysite, which may have been originated from volcanic materials.

(4) Kibushi-clay generally occurs accompanied by lignite seams. It is composed mainly of disordered kaolinite and fine-grained quartz accompanying montmorillonite and small amount of illite.

It is inferred that these clay minerals are mostly detrital derived from weathered granite, argillaceous rocks of the Miocene group and other basement rocks.

And kaolinite crystals in kibushi-clay seem to have grown to some extent probably affected by organic acid after its deposition.

(5) All of these fireclay deposits are intercalated in the Tokiguchi formation of earlier Pliocene age. This formation appears to have been deposited separately in several basins. And its sedimentary features in each basin are not always similar, and appear to reflect the basement geology closely.

Gaerome-clay deposits were developed mostly in Toki basin. This occurrence must probably be in close relation to the granitic rocks distributed widely at Toki area.

Kibushi-clay deposits mainly occur at a few sub-basins of Tajime area, but are scarsely developed at Toki basin. These sub-basins, the basement of which consist mainly of the Palaeozoic rocks, are of very complex shape and very undulated. And so it is inferred that dense drainages gave rise to heavy vegatation at all three sub-basins of Tajimi area. Such complicated reliefs probably promoted formation of swamps, which are favourable for deposition of lignite seams and kibushi-clay beds.

Localization of "kaolin" deposits also seem to be in close relation with depositional environments. In the relatively extensive basins such as Toki, slow sedimentation probably caused mixing of volcanic and detrital materials. Thus good quality "kaolin" with little impurities occurs scarcely in Toki area. And it is assumed that formation of good quality "kaolin" deposit must be intimately related with acidity of lake water.

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# 岐阜県多治見・土岐地方の耐火粘土鉱床の成因

藤井 紀之

## 要 旨

木節粘土および蛙目粘土は本邦の代表的な耐火粘土として知られており，多治見・土岐地方は瀬戸地方などとともにその主産地である。当地方にはこの他に“カオリン”と称される白色粘土が賦存し，おもに白色磁器の原料として採掘されている。これらの粘土は，いずれも下部鮮新統の土岐口累層中に挟在しているが，地質的産状，鉱物組成，主成分であるカオリン鉱物の種類，耐火度などの多くの点において，それぞれ異なる特徴を有している。この研究においては，このような事実を基として各種の耐火粘土鉱床の形成過程について考察した。

当地方の土岐口累層は，多治見盆地と土岐盆地に分かれて堆積したと考えられる。多治見盆地の基盤は大部分古生層からなつており，小名田，大畑，大洞など幾つかの小盆地が相互に連繋し，連珠状の複雑な形の堆積盆地を形成している。一方土岐盆地の基盤は大部分が中新統と花崗岩類である。そして多治見盆地とは対照的に，広くしかも屈曲の少ない形状を呈している。なお土岐口累層は，土岐累層と呼ばれる厚い砂礫層（上部鮮新統）によつて広くおおわれている。

いわゆる“カオリン”はおもに大洞地区に産出する。1〜2の特定の層準に薄層をなして断続し，特徴的な白色を呈することによつて他の粘土と区別される。一部で粗粒の凝灰岩と漸移すること，鏡下で酸性凝灰岩の組織が認められることなどの点から，凝灰岩から自生的に生成されたものと推定される。主として径0.1〜0.3$\mu$の木片状のハロイサイト（メタハロイサイトを混じえる）からなり，少量の石英および火山ガラスを伴つている。

蛙目粘土の場合は，大部分が土岐盆地に分布している。土岐口累層の基底部に比較的厚層をなして賦存し，粘土の他に石英や長石などの花崗岩質の鉱物を多く含む所から，風化・分解された花崗岩に由来する物質が余り淘汰を受けずに原地から近い所に堆積したものと考えられる。粘土鉱物としては，主としてカオリナイトを含む場合とハロイサイト（一部脱水したものが多い）を多く含む場合とがあり，それは蛙目粘土の淘汰の程度と密接な関係がある。すなわち粘土分の多い試料程カオリナイトを多く含み，粘土分の少ないものにはハロイサイトが多く含まれるという傾向が明瞭に認められた。ちなみにカオリナイトは径0.1$\mu$前後の不規則な板状のもで，ハロイサイトは長径0,5$\mu$程度で管状または長柱状を呈するものが多い。このような粒度の違いが両者の分別を促進したものと推定される。

木節粘土は軟質で可塑性に富み，常に亜炭層に伴つて賦存する。おもに多治見盆地によく発達している。通常カオリナイトと細粒の石英からなり，モンモリロナイトおよび少量のイライトを随伴している。このように同じ場所で，しかも同一の地層に挟在するにもかかわらず木節粘土と“カオリン”の鉱物組成がまつたく異なつていることは，両者の原物質が異なることを示すものである。“カオリン”が凝灰岩に由来する自生質の粘土鉱物からなるものである以上，木節粘土の起源は基盤岩類に由来する砕屑質の物質に求めるのが自然であろう。

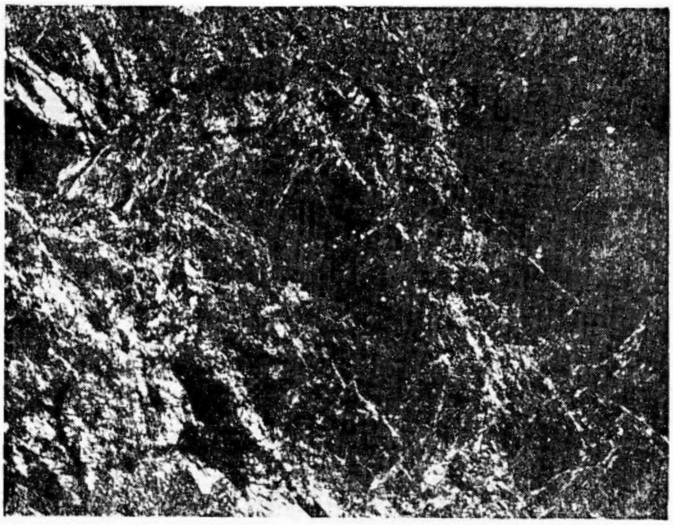
最近の古植物学的資料によれば，土岐口累層の堆積当時は亜熱帯性の気候条件下にあつたと考えらており，基盤岩類が著しい風化を受けた跡が土岐口累層におおわれて保存されている状況も随所に認められる。土岐市南端の柿野カオリンも，当時花崗岩類の風化によつて生成されたと推定される現地残留性の鉱床であるが，カオリン鉱物としては長柱状のハロイサイトと板状のカオリナイトの双方を含んでいる。このような事実は，蛙目粘土が風化花崗岩に由来するという前述の推定を裏付けるものである。木節粘土の場合も，カオリナイトは主として基盤の花崗岩からもたらされたと思われる。また中新統の粘土質岩の影響も無視できない。モンモリロナイトおよび一部のカオリナイトは，中新統

の岩層に由来するものである可能性が強い。なお木節粘土の場合，カオリナイトの定方位集合体が樹枝状，網目状などの組織を形成していることが多く，これは木節粘土の堆積後に有機物などの影響でカオリナイトが成長したことを示している。しかし基盤花崗岩の風化状態から見ても，カオリン鉱物の基本構造は堆積前に形成されていたと考えるのが妥当である。

これら各種の耐火粘土は，地域的にかなりかたよつた分布を示す。これは各堆積盆地の基盤の違いと，土岐口累層の堆積過程の相違によつて生じたものと思われる。蛙目粘土が土岐盆地に大量に分布しているのは，盆地の基盤が花崗岩類と中新統からなるという事実から容易に説明できる。また多治見盆地の地形が複雑な起状に富んでいたことは，土岐口累層の堆積途中に多くの小規模な沼沢地が形成される要因になつたと考えられる。これらの沼沢地が亜炭および木節粘土の集積し易い環境を形成したものであろう。また土岐盆地には凝灰岩や凝灰質粘土層は発達しているが，良質の“カオリン”はほとんど産出しない。これは堆積盆地の広さや堆積速度の関係で，多くの砕屑物質が混入したこと，亜炭層の発達が乏しくしたがつて湖水の酸度も多治見盆地とはかなり違つていたことなどの理由によるものと推定される。

# PLATES
# AND
# EXPLANATIONS

(with 10 Plates)

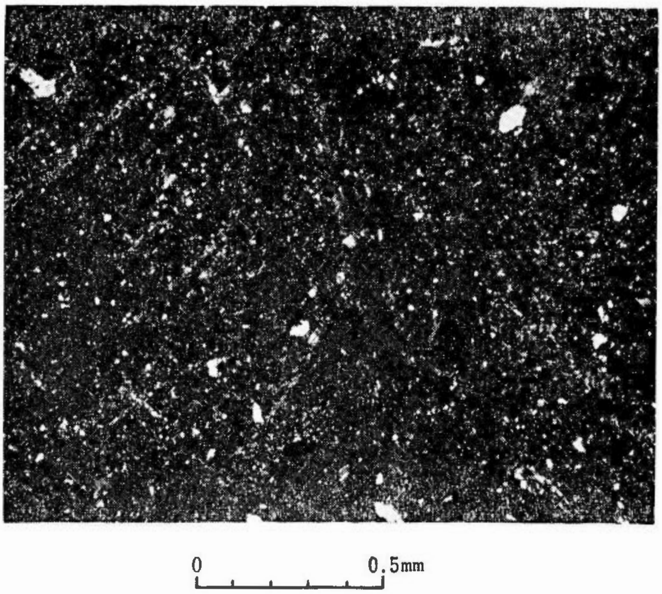


0 0.5mm

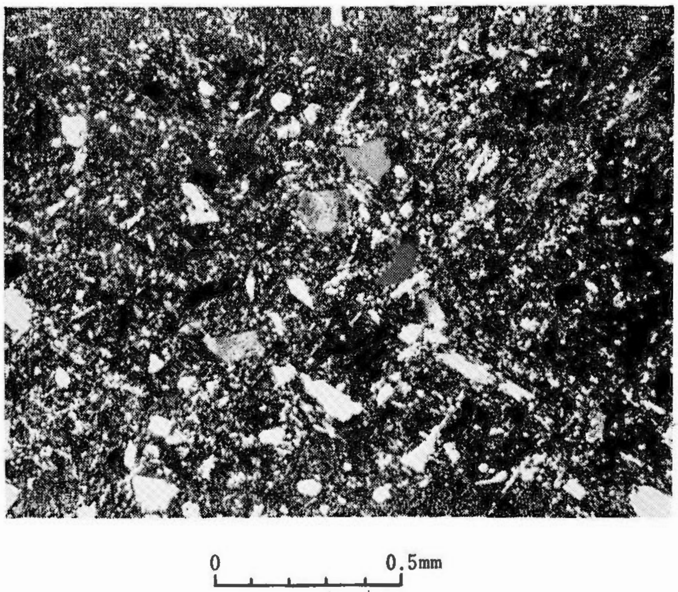
1. Mesh structure in kibushi-clay (O-42 from Tado workings, Obora) (crossed nicols)

0 0.5mm

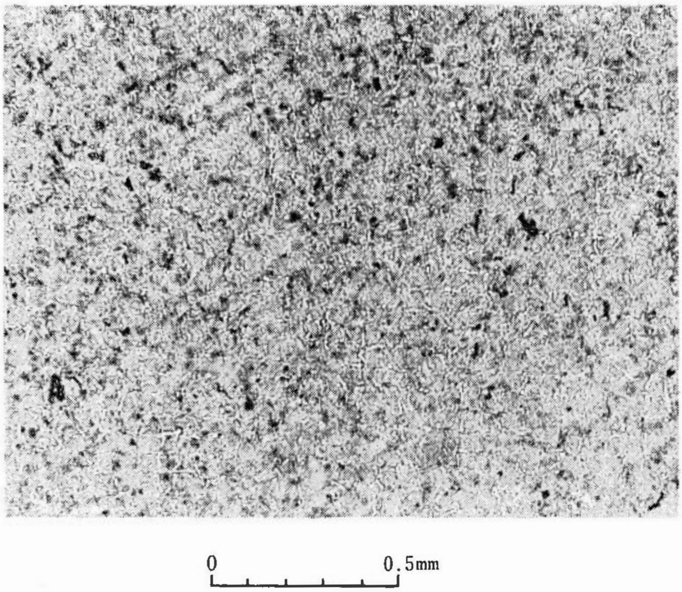
2. Fibrous structure in kibushi-clay (O-94 from Moriyama workings, Obora) (crossed nicols)



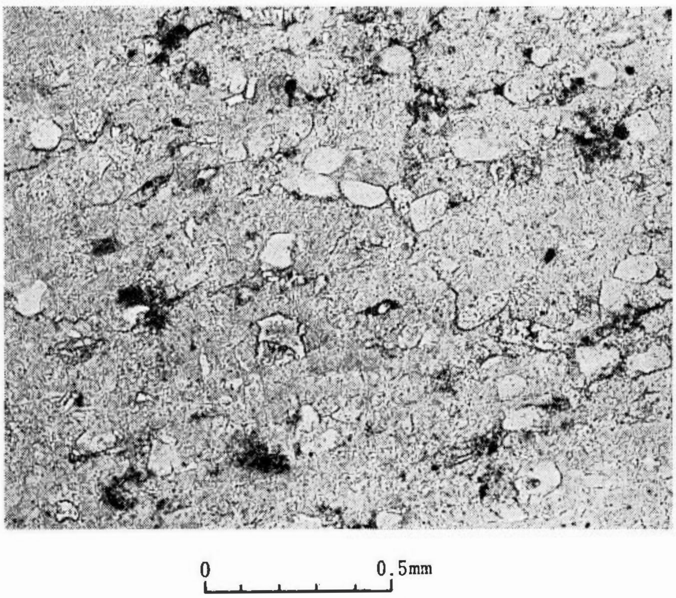
1. Microphotograph of miscellaneous clay (N-93 from Aisei workings, Onada) (crossed nicols)



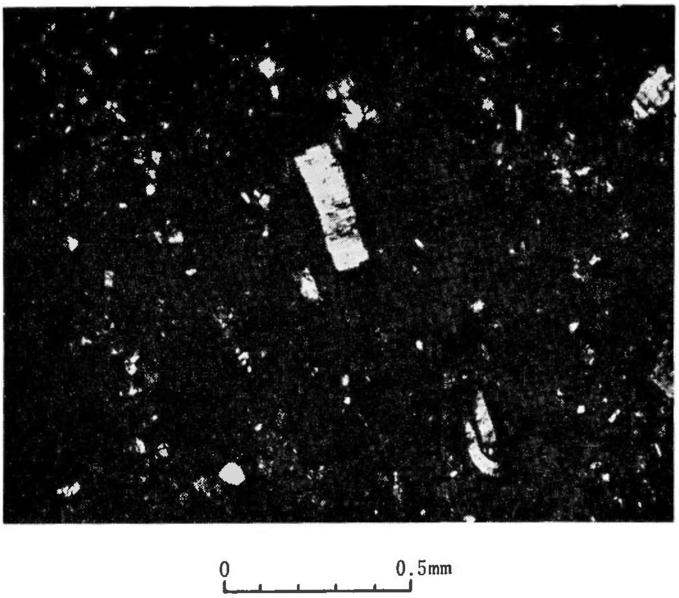
2. Microphotograph of gaerome-clay (T-37 from Sengen workings, Toki) (crossed nicols)



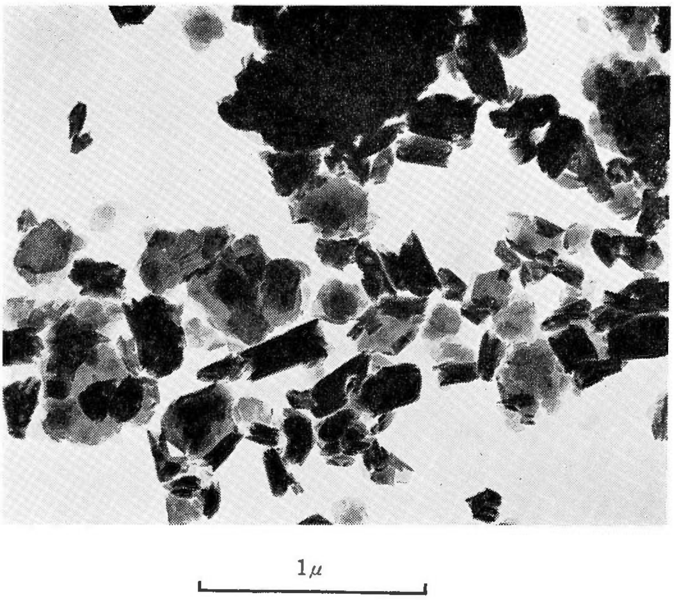

1. Fine-grained glass shards in tuff O-83 from Katahira workings, Obora)



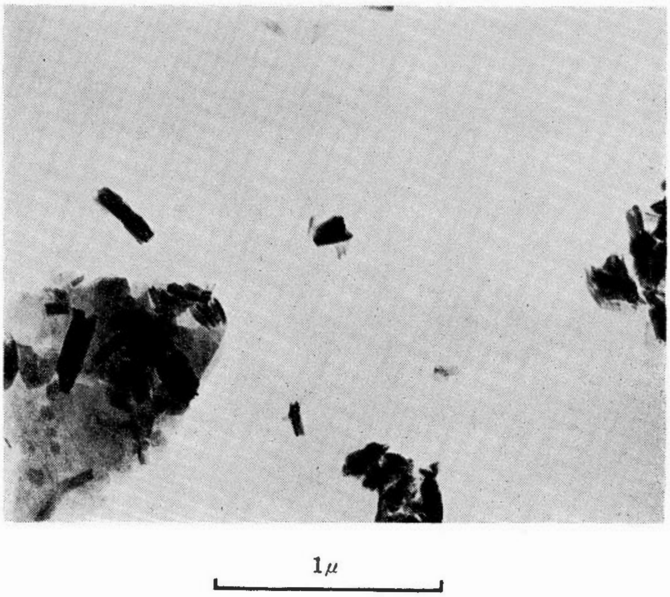

2. Glass rings in the so-called kaolin (O-97 from the place 300 m east of Watanabe workings, Obora)



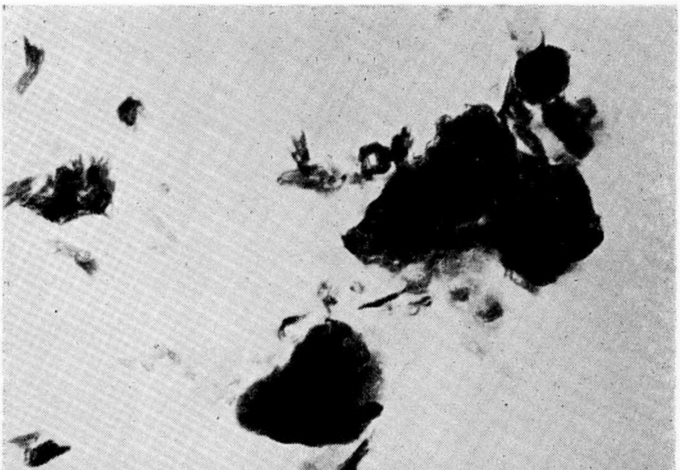

1. Biotite pseudomorph composed of feather-like kaolin minerals in the so-called kaolin (O-4 from Nakayama workings, Obora) (crossed nicols)



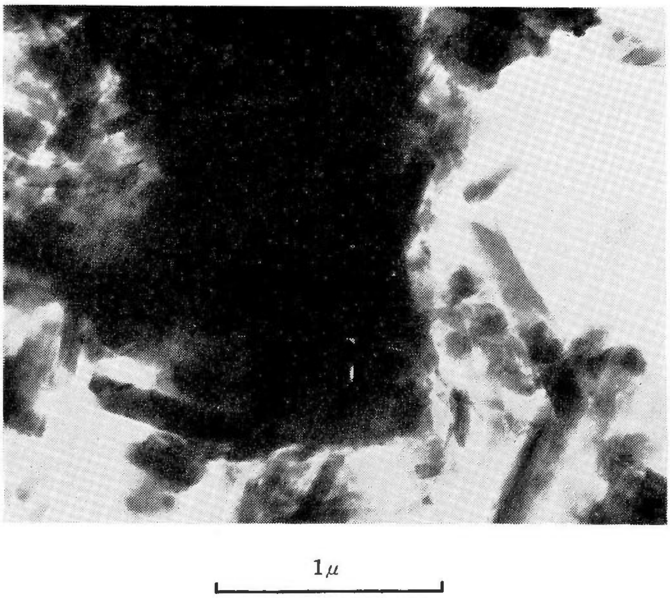

2. Lath shaped crystals of halloysite in "kaolin" (O-8 from Meiho workings, Obora)



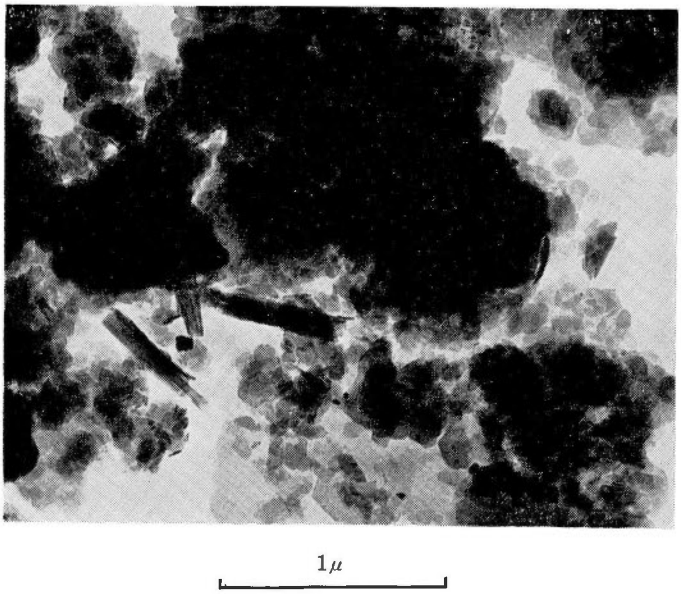
1. Lath shaped crystals of halloysite in "kaolin" (O-19 from Nishiyama workings, Obora)



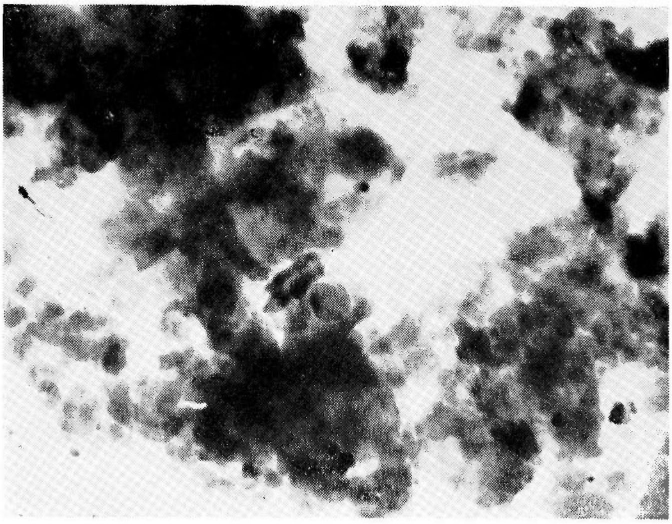
2. Lath and tube shaped crystals of halloysite in "kaolin" (T-81 from Oizawa, Toki)



1. Tubular crystals of halloysite in washed gaerome-clay (T-59 from Sawabora workins, Toki)

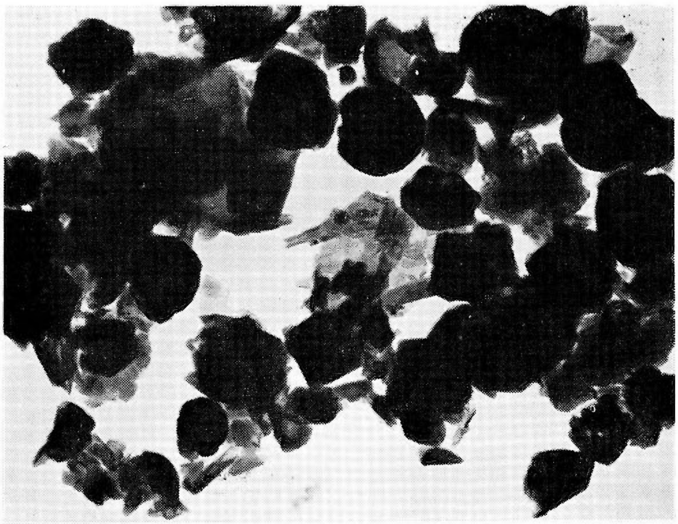


2. Six-sided crystals of kaolinite in washed gaerome-clay (T-37 from Sengen workings, Tokyi)



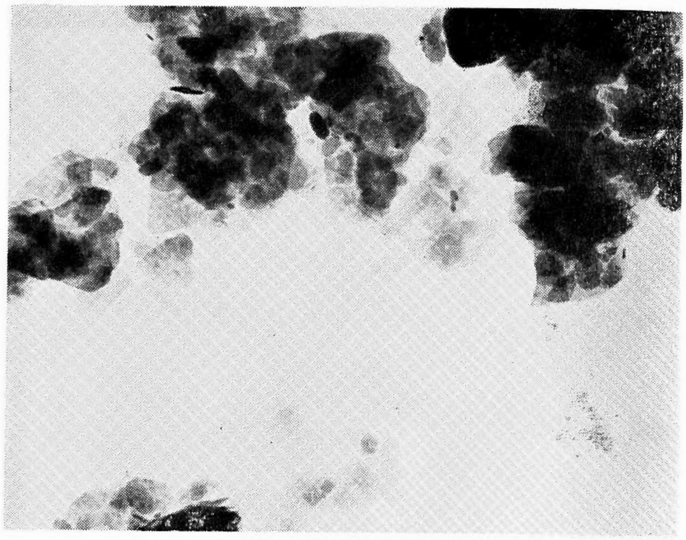
1μ

1. Six-sided crystals of kaolinite in washed fine sand (T-20 from Kinrei workings, Toki)



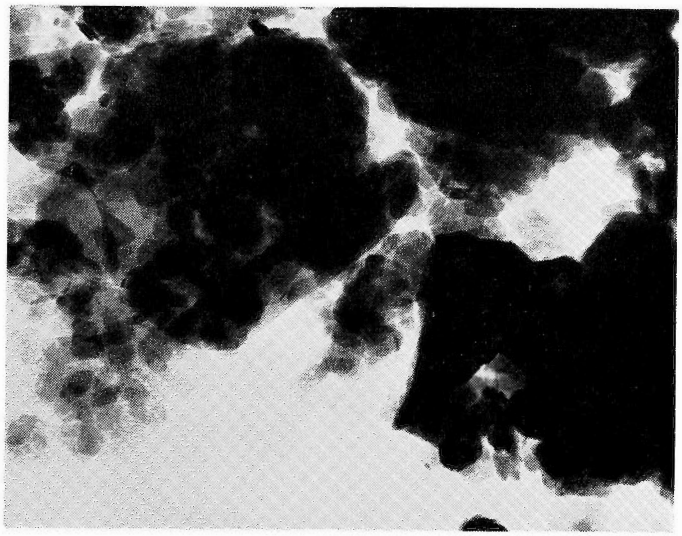
1μ

2. Globular crystals of halloysite in washed gearome-clay (O-70 from Yamaso workings, Obora)



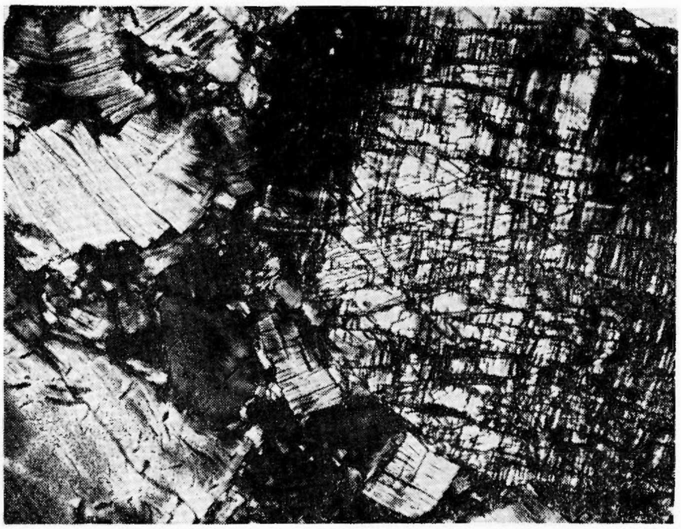
1μ

1. Six-sided crystals of kaolinite in kibushi-clay (O-42 from Tado workings, Obora)



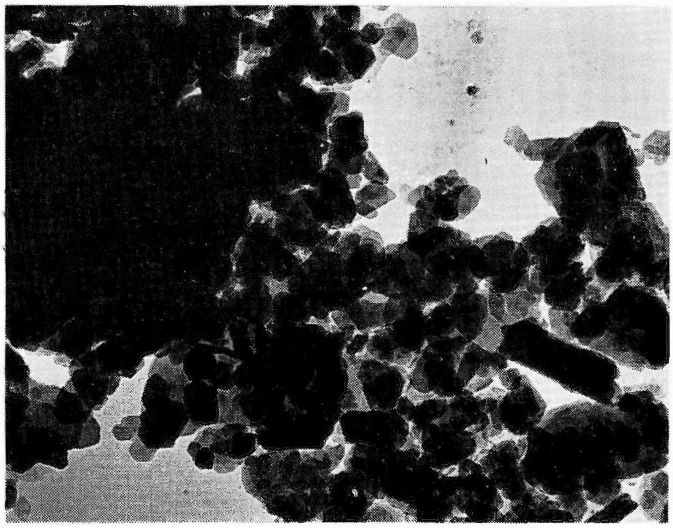
1μ

2. Six-sided crystals of kaolinite in kibushi-clay (N-98 from Aisei workings, Onada)



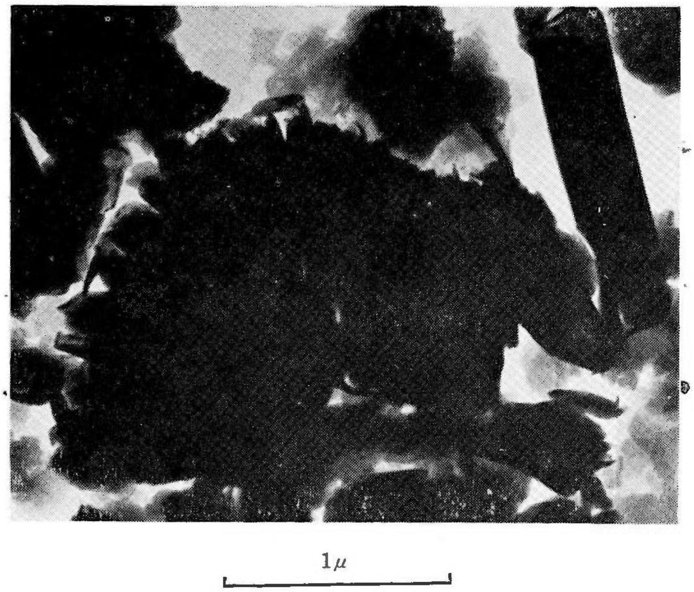
0 0.5mm

1. Biotite pseudomorphs of kaolin minerals and K-feldspar with microcline structure in weathered granite, Kakino mine (crossed nicols)

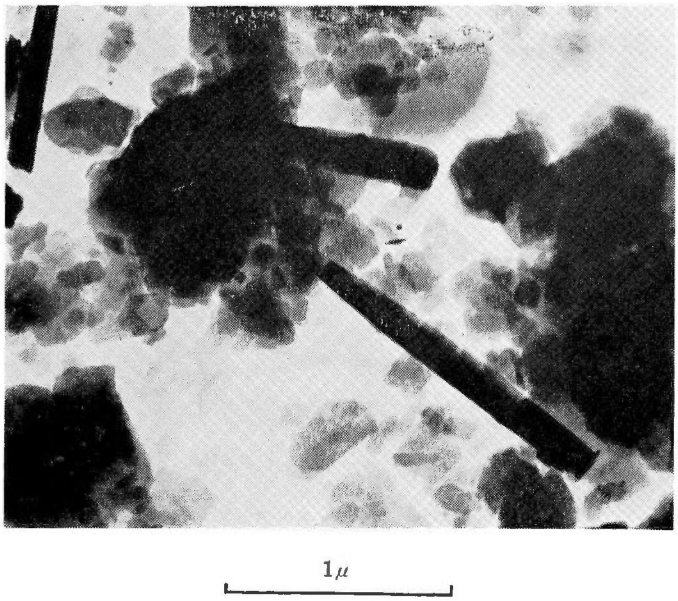


$1\mu$

2. Distinctly six-sided kaolinite crystals accompanying some tubular metahalloysite in washed gaerome-clay, Kakino mine



1. Tubular crystals and feather-like book of kaolin minerals in washed residual-kaolin, Kakino mine



2. Six-sided crystals of kaolinite and tubular crystals of meta-halloysite in washed residual-kaolin, Kakino mine

地質調査所報告は1報文について報告1冊を原則とし，その分類の便宜のために，次のようにアルファベットによる略号をつける。

A．地質およびその基礎科学に関するもの
- a．地　質
- b．岩石・鉱物
- c．古生物
- d．火山・温泉
- e．地球物理
- f．地球化学

B．応用地質に関するもの
- a．鉱　床
- b．石　炭
- c．石油・天然ガス
- d．地下水
- e．農林地質・土木地質
- f．物理探鉱・化学探鉱および試錐

C．その他

D．事業報告

As a general rule, each issue of the Report, Geological Survey of Japan will have one number, and for convenience's sake, the following classification according to the field of interest will be indicated on each Report.

A. Geological & allied sciences
- a. Geology
- b. Petrology and Mineralogy
- c. Paleontology
- d. Volcanology and Hot spring
- e. Geophysics
- f. Geochemistry

B. Applied geology
- a. Ore deposits
- b. Coal
- c. Petroleum and Natural gas
- d. Underground water
- e. Agricultural geology and Engineering geology
- f. Physical prospecting, Chemical prospecting & Boring

C. Miscellaneous

D. Annual Report of Progress

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FUJII, N.

**Genesis of the Fireclay Deposits in Tajimi-Toki District, Gifu Prefecture, Central Japan**

Noriyuki FUJII
地質調査所報告 No. 230, p. 1~56, 1968
34 illus., 10 pl., 5 tab.

A few kinds of fireclay deposits, which are kibushi-clay, gaerome-clay and the so-called kaolin, are developed in the earlier Pliocene Tokiguchi formation of this district. Each of these fireclays has its own characteristics concerning fabric, mineral composition, type of main kaolin mineral and so on. In the present paper, geological occurrences and mineral composition of these clays are described. And it is discussed that the differences among their characteristics are owing to the differences in genesis. Saying shortly, kibushi- and gaerome-clays are of detrital origin, and "kaolin" may be authigenic.

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