Origin of the gross morphology and internal texture of tufas of Shirokawa Town, Ehime Prefecture, southwest Japan

Akihiro Kano* and
Hidenori Fujii**

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* Department of Earth and Planetary Systems
Science, Graduate School of Science,
Hiroshima University, Higashi-hiroshima
739-8526, Japan

** Nippon Total Science Co. Ltd., Minoshima-
cho, Fukuyama 721-0957, Japan

Abstract

Gross morphology and internal texture of tufa deposits in Shirokawa Town, Ehime Prefecture, vary due to local environments (including flow condition, substrate topography, and associated biota) within the tufa-bearing stream. Variations in gross morphologies of the tufa deposits include 1) tufa encrusting boulders, 2) plane slope, 3) mound, 4) terraces, and 5) cascade. These types are basically related to variations in the original topography although they share similarities in a local environment of strong water flow which is an essential condition for active tufa deposition. Laminated internal texture, the most striking feature of the Shirokawa tufa, normally develops in tufas colonized by cyanobacteria inhabiting the surface. Regular lamination is most commonly developed along the narrow water passage of the lower stream where strong water flow continues throughout the year. The laminated tufas consist of repetition of dense lighter-colored laminae and porous darker-colored laminae, which correspond to the summer and winter deposits, respectively. The lighter laminae are characterized by thick calcite encrustation on upward-growing filamentous cyanobacteria. The darker laminae also mainly consist of calcite encrustation of cyanobacterial filaments, but the porosity largely remains. The annual rhythm seen in the laminated texture can be explained by seasonal variation in abiotic calcite precipitation rate (large in summer and small in winter) which increases with water temperature, Ca^{2+} contents, and flow strength. Because of the well-defined chronological constraints, continuous deposits of laminated tufas can be excellent material for reconstructing terrestrial climates.

Key words : tufa, travertine, freshwater carbonate, gross morphology, laminated texture, cyanobacteria

Introduction

A tufa is a form of freshwater carbonates deposited by physico-chemical and biological processes in open-air conditions. Tufa-depositing water normally originates from an underground water system of limestone and supersaturated with respect to calcite (Kano et al., 1998), and it should be in ambient temperature (close to the annual average temperature of the locality). Similar carbonate can be deposited from water of hot springs, however they are called as travertines according to the recent definition of Ford and Pedley (1996).

Tufas display a wide variety of structures in both terms of gross morphology and internal texture. The variety can be seen even in a single tufa-depositing stream. Several previous works, mainly on European and North American tufas, have discussed possible

controls of the structures of tufas. The gross morphology of tufas varies with local conditions of topography and hydrology (Pedley, 1990). The internal textures are largely associated with biota colonizing on the tufa surface (Chafetz and Folk, 1984), and with seasonal change in weather conditions (e.g., Yoshimura et al., 1996a). Additionally, tufas often form laminated internal texture. The lamination normally shows regular rhythm which results from seasonal change in developmental conditions, as tree rings of woods (Chafetz and Folk, 1984; Kano, 1997). These facts have stimulated our interests in tufas because structures of tufas record paleoenvironmental conditions, and because the laminated texture of tufas provides well-defined chronological constraints for paleoclimatological analyses.

Since the first description of tufas in Japan by

Town, Ehime Prefecture (Fig. 1 a). It is so far one of the largest tufa deposits of Japan. The tufa deposits are exposed along the stream of 460 m long and 120 m in altitude difference. Hydrologic and chemical conditions of the water were described and discussed in the previous paper (Kano et al., 1999 b). They are summarized as follows.

The tufa-bearing stream receives water from seven springs. It is divided into the upper stream and the lower stream at the spring 2 (Sp-2; Fig. 1 b). In the upper stream, water first flows on an artificial narrow passage from St 29.0 to St. 26.3, and then spreads on a wide passage. During the periods of low flow, water mostly soaks into the tufa deposits on the wide passage. From the spring 2, the water continuously flows down along a narrow passage of the lower stream.

The water from the uppermost and largest spring (Sp-1) is groundwater in origin, coursing through an underground water system. It records a small seasonal variation in temperature ranging from 13 to 15°C, and is rich in dissolved Ca^{2+} (about 1.5 mM; Kano et al., 1999 b). When the water issues, it is slightly supersaturated with respect to calcite, and it has much higher partial pressure of CO_2 (Pco_2) than the atmosphere. Temperature of the streamwater varies and approaches to air temperature as it flows away from the orifice. Pco_2 of the water decreases rapidly during the first 50 m-flowing from the orifice, and decreases slowly in the lower stream. Together with CO_2 degassing, pH increases downcurrent. Then, the water becomes increasingly supersaturated with respect to calcite. Gradual decrease in Ca^{2+} content and in alkalinity (carbonate alkalinity) to downstream indicates that the water deposits CaCO_3 during flowing on the water passage. The water of the stream normally satisfy the chemical conditions for tufa deposition (Kano et al., 1998); Ca^{2+} content is more than 1.2 m M and saturation index for calcite is more than 0.5 (Kano et al., 1999 b).

Chemical and hydrological conditions vary throughout a year. Alkalinity and Ca^{2+} content of the spring water are high in summer and autumn that is probably associated with evaporation of water in underground and active dissolution of limestone in summer due to high Pco_2 in soil air. Amount of water flow varies generally following the change in rainfall, which is observed at the nearest weather station (Chikanaga, Ehime Prefecture). Depositional rate of tufa, calculated based on chemical (alkalinity) and hydrologic (amount of water flow) data, is in higher values in summer (Kano et al., 1999 b).

Fig. 1 b shows the distribution of tufa deposits along the stream. The upper stream develops continuous and thick (more than 100 cm) tufas, whereas tufa development of the lower stream is discontinuous and thin (normally less than 50 cm; Kano et al., 1999 b). The locations without tufa development are generally

characterized by stagnant flow condition, such as, small pools of 1~5 m in diameter and more than 30 cm in water depth. In these locations, pisoids are often found.

Methods

We had several occasions to observe and collect of tufa deposits since May 1995. For mapping the stream, we used 29 stations (Fig. 1 b) set by Dr. Y. Inokura (Kagoshima Univ.); distance between two stations nearby is 11~20 m. In order to indicate a precise location, numbers with a decimal fraction of the station (e.g., St 29.0) were used in the text. Tufa deposits were collected by cutting with an ordinary saw. In order to monitor development of laminated texture, series of sampling were done from a small portion (the area is less than 30 cm in diameter) of the same tufa deposit at St 9.2.

Organisms were collected from surfaces of some laminated tufas. They were grown on 1.5% agar plates with culture medium (MDM-medium; Nishizawa and Chihara, 1979) in a petridish. A few weeks later, the agar plates were observed under an optical microscope. The recognized organisms were filamentous cyanobacteria and unicellular organisms (algae or cyanobacteria). Then, the filamentous cyanobacteria was isolated by using a sterilized needle and cultured in an Erlenmeyer flask with the culture medium and a cotton seal.

The specimens of tufas and cultured cyanobacteria were observed under optical, fluorescence, differential interference, and scanning electron microscopes. Generally, a lamination is best and easily demonstrated in a view obtained using a scanner (shown in Fig. 6 a). Because the tufas are fragile, the specimens were first hardened by resin diluted with acetone, and then made into thin sections.

Some of the specimens with laminated internal texture were used for analyzing porosity and content of insoluble residue. The specimens were dried in room temperature and cut into cuboids. Z-axis of a cuboid is perpendicular to the laminae and area of the x-y rectangular is approximately $2 \times 3 \text{ cm}^2$. Then, the cuboid was divided at the lamina boundaries. Volume of each divided specimen (called as sub-specimen) is known by lengths along x, y, and z-axes. The sub-specimens were dried at 110°C for twelve hours, weighed, dissolved in diluted (5%) acetic acid, and then passed through a $0.2 \mu\text{m}$ filter. The filter was dried and weighted. The weight increase of the filter after the filtration was adopted as weight of insoluble residue. The mineralogy of the insoluble residue was analyzed by x-ray diffractometer. It mainly consists of quartz and clay minerals (smectite and kaorinite) with minor contents of organic matter.

Porosity of each sub-specimen (P) was calculated from total volume (V), total weight (W_T) and weight of

Fig. 2. Schematic sketches of the five gross morphological types of the tufas in Shirokawa Town.

insoluble residue (W_{IR}) and by employing densities of calcite (2.71 g/cm^3) and quartz (2.65 g/cm^3) for the soluble and insoluble components, respectively.

$$P (\%) = 100 - 100 \{ (W_T - W_{IR}) / 2.71 + W_{IR} / 2.65 \} / V$$

Gross morphology of the tufas of Shirokawa

1. Classification of gross morphological types

Modes of previous classifications of tufa deposits are diverse. Classifications of Schneider et al. (1983) and Pentecost and Lord (1988) focused biological char-

acteristics and named the deposits by means of associated organisms (e.g., Oscillatoriacean tufa and bryophyte tufa). Ordóñez and García del Cura (1983) proposed sedimentological terminology, such as stromatolitic tufa and detrital tufa. Pedley (1990) presented several depositional models of tufas and covered a large variety in depositional settings (rivers, hill slopes, lakes, and so on). Although his work is extensive, none of his specified models fits to the case of Shirokawa. The tufa-depositing stream of

Fig. 3. Tufas, paleo-tufa, and pisoids of Shirokawa. (a) A tufa encrusting boulders at St 3.5. (b) A mound type tufa at St 14.5. The surface is colonized by mosses and cyanobacteria. (c) Mound and pools around St 15.0. (d) Terraces at St 24.8. (e) A cascade type tufa around St 13.0. (f) A paleo-tufa at St 10.1 showing traces of roots (R). Diameter of the lens cap is 7.5 cm. (g) Pisoids (P) at St 26.1. (h) Thin section of a pisoid collected at St 26.1. It consists of the sandstone nucleus (Ss) and the laminated cortex.

Shirokawa represents characteristics of two of Pedley's (1990) perched springline model (hill slopes) and fluvial model (rivers).

Because no previous classification efficiently covers a variety of the tufa deposits of Shirokawa, we propose a classification in our own scope. Based on the gross morphology, the tufa deposits are classified into the following five types; a) tufa encrusting boulders, b) plane slope, c) mound, d) terraces, and e) cascade (Fig. 2). We are fully cognizant of great variety of gross morphology place to place. Therefore, this classification is somehow a broad generalization. It should be mentioned that some deposits exhibit combined characteristics of two or more types.

The five types of gross morphology are described below, with their schematic sketches (Fig. 2) and photographs (Figs. 3a~e).

(1) **Tufa encrusting boulders**: Numerous limestone boulders occur on the tufa-bearing stream. Tufa commonly encrusts the upper surface and downstream side of these boulders (Figs. 2a and 3a). This type of tufa is normally thin (less than 20 cm) and the substrates partly remain uncovered. It mainly occurs in locations downstream St 14.5. Cyanobacteria is the most common biota on the surface of tufa and moss is associated.

(2) **Plane slope**: The tufa forms a plane slope to the downstream declining more than 5 degrees at St 7.2~7.4, St 19.0~19.7, St 21.2~21.5, and several locations of the upper stream. Substrates of this type are mudstone bedrock (Fig. 2b), paleo-tufa, and artificial water passage (St 26.3~28.7). Outcrops along the stream expose mudstone and sandstone of the upper Jurassic-lower Cretaceous Torinosu Group. Although these strata generally dip 10~20 degrees to the north (upstream; Kano et al., 1999b), the mudstone beds, in some places, form a plane substrate declining several degrees to the downstream. Tufas of this type are generally thin (less than 30 cm) in the lower stream, but thickly (more than 1 m) developed on the western tributary of the upper stream (see Fig. 1b). Cyanobacteria is dominant on the surface of the slopes. However, moss commonly inhabits slopes of the upper stream where water is failed during the periods of low flow.

(3) **Mound**: Mound type is the most common gross morphology in the lower stream, especially in the narrow water passage between St 14.5 and 18.7. The mounds are characterized by a tongue-like shape protruding to the downstream direction (Fig. 3b), and the base of the tongues normally above the stream water level (Figs. 2c and 3b). They mostly appear as small-scale dams (less than 100 cm in height), associated with pools forward and backward (Figs. 2c and 3c). This probably results from upward and forward growth of the mounds. Limestone boulders commonly occur as substrates of the mounds. Cyanobacteria

is common on the surface, and moss inhabits the surface parts temporally retreated by water.

Kano (1997) used 'cascade tufa type' for describing this type of tufa. However, following other previous descriptions (e.g., Pedley, 1990), the term 'cascade' should be used for a deposit forming a waterfall that is much higher than the mounds.

(4) **Terraces**: Terraces are observed only in the widely (about 5 m) spread passage at St 24.4~25.0. Here, a number of terraces display a stairs-like relief (Figs. 2d and 3d). A single terrace, 30~70 cm in height and 20~100 cm in width, consists of a raised rim around the edge and a slightly depressed plane in backward. The gross morphology is similar to rimstone seen in a limestone cave, and is formed due to the precipitation of calcite as the water flows over the edge and down the vertical side to the next terrace below. The tufa deposits are thick (more than 180 cm), and so the substrate condition is not exposed. The terraces are covered by relatively weak water flow, and even temporally retreated by water during periods of low flow. Cyanobacteria is common, but moss is more abundant on the surface, probably due to weak water flow and relatively strong light intensity.

(5) **Cascade**: Small-scale waterfalls covered with tufa deposits are called as cascades. They occur near St 11.0, 13.0 (Fig. 3e), 21.5 and along the water from Sp-5 (see Fig. 1b). Height of these cascades reaches 8 m. Tufa deposits on the cascades can be thick (more than one meter). They form at previous sites of waterfalls (Fig. 2e), or where boulders protrude above the water level. Cyanobacteria and moss are found on the surface.

2. Remarks of other calcareous deposits

(1) **Paleo-tufa**: Other than currently depositing tufas, paleo-tufas are exposed along the stream. The paleo-tufa is here defined one which used to have been deposited and now covered by a soil layer. Because soil covers the upper surface, gross morphology of the paleo-tufas is hardly recognized.

Deposition of the paleo-tufas was ceased by shift of water passage. Considering the large depositional rate of tufas, change in the topography could shift water passage in a short time period. Therefore, the paleo-tufas are probably not very old. Age of deposition may be younger than Quaternary although any dating has not been done yet.

Fig. 1b shows distribution of the paleo-tufas. In the lower stream, they occur most largely near St 9.4~11.0 (Fig. 3f) and St 12.0~13.7. In the upper stream, the paleo-tufas are ubiquitously distributed between the three water passages and form topographic rises. This occurrence indicates that water had changed the passage as the tufa deposition developed the topographic rises.

The exposed sections of paleo-tufas display internal textures. The outcrop beside St 23.0 (collapsed tufa in

Fig. 4. Vertical sections of tufa specimens. (a) A laminated tufa in field (St 9.2). Water flows from left to right (as well for (b)–(d)). (b) A laminated tufa showing an unconformity surface (St 9.2). Thickness of each lamina decreases on the downstream flank and overhanging surface. (c) A laminated tufa collected from St 6.5. Development of lamination is limited in the uppermost 6 cm. (d) A tufa colonized by mosses (St 3.0). Traces of rhizoid disturb development of laminated texture.

Fig. 1 b) shows continuous several hundreds laminae. Traces (void space) of roots very commonly occur in the paleo-tufa (Fig. 3 f).

(2) **Pisoids** : Pisoids, several mm to 10 cm in diameter, normally occur in weakly flowing water (Fig. 3 g). They consist of nuclei encrusted by calcite cortex. Color of the surface is white to light gray, showing that cyanobacteria does not sufficiently colonizes the surface. The nuclei are mainly gravel of mudstone, sandstone, and limestone, but can be a nail and a fragment of wood. Thickness of cortex ranges from less than 1 mm to several cm. A thicker cortex commonly consists of very fine (less than 0.5 mm in thickness) laminae occasionally forming knobby texture (Fig. 3 h).

Laminated internal texture of tufas of Shirokawa

The tufas of Shirokawa generally develop laminated internal texture (Takebe et al., 1996). However, in locations temporally retreated by water (e.g., the slope between St 25.0 and St 21.0), the tufas are intermittently deposited and cannot develop a clear

lamination. On the contrary, the clear lamination tends to be developed in tufas covered permanently with water flow and often associated with cyanobacteria. These conditions are commonly adopted for the tufas in the lower stream, but not in the upper stream.

1. Characteristics of the laminated texture

(1) **Textures in hand specimens** : The laminated texture consists of repetition of darker-colored laminae and lighter-colored laminae (Fig. 4). The boundary between laminae is normally a well-defined smooth surface, but in some cases weakly ragged. Many of the specimens limit the development of laminae in a superficial part less than 20 cm in thickness, and the number of laminae rarely exceeds 100 (Figs. 4 b and c). The core (deeper part) of tufas appears traces of root with diameter of 1~10 mm (Fig. 4 c). In some specimens, irregularity of the lamination indicates discontinuous deposition. For instance the specimen shown in Fig. 4 b exhibit unconformable pattern of lamination.

Thickness of a single lamina mostly ranges from 0.4

Fig. 5. Results of analyses of sub-specimens (laminae). Specimens used for the analysis are the mound and plane slope types collected at St 7.8, 12.0, 15.2 and 17.8. (a) Porosity of the sub-specimens of lighter-colored and darker-colored laminae. (b) Weight percentage of insoluble residue contents. (c) Porosity and insoluble residue contents of the sub-specimen of the tufa collected from St 17.8 in March, 1996. The sub-specimen of 16.4~26.2 mm from the surface consists of two darker laminae and a lighter laminae. (d) Annual growth rate of the analyzed tufa deposits. (e) Annual calcite accumulation rate of the analyzed tufa deposits.

Table 1. Average values of porosity (%), contents of insoluble residue (IR ; %), annual growth rate (GR ; mm), and annual calcite accumulation rate (CA ; grams in CaCO₃) of four analyses laminates tufas collected at St 7.8, 12.0, 15.2 and 17.8.

Locality	Porosity (%)	IR (%)	GR (mm)	CA (g/cm ²)
7.8	62.7	7.1	3.2	0.30
12.0	66.5	5.6	3.0	0.28
15.2	65.6	11.7	3.0	0.24
17.8	71.7	9.6	5.0	0.33

to 4 mm and varies with localities, and even within a samples. The specimens of mound type tufas show the thickest development of laminae on the top, and the topmost laminae thin out toward the overhanging surface (Figs. 4 b and c). The tufas colonized by moss may have lamination, but it is obscure and disturbed by traces of rhizoid (Fig. 4 d).

(2) **Porosity and insoluble residue** : Porosity and contents of insoluble residue differs due to the lamina type. The darker laminae are more porous (porosity ranges from 62 to 79% ; 71% in average) than the lighter laminae (porosity ; 48~73%, 61% in average ; Fig. 5 a). Contents of insoluble residue are slightly higher in the darker laminae (4~15% in weight ; 9% in average) than the lighter laminae (4~14%, 7.5% in average ; Fig. 5 b). These tendency can be seen within a single tufa deposit (the tufa at St 17.8 ; Fig. 5 c).

Average values of porosity and contents of insoluble residue are different among the four analyzed tufas (Table 1). The tufa at St 17.8 is more porous than other three tufas. The largest contents of insoluble residue are recorded in the tufas at 15.2 (Table 1).

(3) **Growth** : Fig. 6 a shows thin-sections of superficial parts of a laminated mound-type tufa at St 9.2 collected between May 1998 and July 1999. From left to right in Fig. 6 a, the surface of tufa changes from a darker lamina (D2) in May 1988, a light lamina (L2) in August 1998, a dark lamina (D1) in October 1988~April 1999, to a light lamina (L1) in July 1999. This indicates that the lighter laminae were deposited in seasons covering summer, and that the darker laminae were deposited in seasons covering winter. These observations strongly confirm previous statement that the lamination of tufas is annual (e.g., Takebe et al., 1996 ; Yoshimura et al., 1996c ; Kano, 1997). For the tufa at St 9.2, depositional season of the lighter laminae starts around July, ends around September, and is probably shorter than depositional season of the darker laminae.

(4) **Growth and accumulation rates** : We regard a pair of adjacent lighter and darker laminae as a one-year deposit, and estimate annual growth rate (Fig. 5 d) by the thickness, and calcite accumulation rate (Fig.

5 e) by weight of the soluble component per unit surface area (cm²) for the analyzed four tufas. Estimation of the values is based on the assumption that lamina boundaries were formed at the same times every year. However, this assumption is incorrect in the strict sense. Therefore, the data of Figs. 5 d and 5 e may involve the deviation causes by variation in depositional periods among the pairs of two adjacent laminae.

The two rates are concentrated in relatively narrow ranges ; the annual growth rate mainly ranges from 2.0 to 4.0 mm (Fig. 5 d), and the annual calcite accumulation rate mainly ranges from 0.15 to 0.40 g/cm² (Fig. 5 e). Difference in the rates among the four tufas is relatively small although the tufa at St 17.8 records sufficiently larger values in the annual growth rate (Table 1).

2. Microtexture

In thin sections, the lamination is recognized by the difference in porosity between darker and lighter laminae (Fig. 6 b). This is consistent with the result of porosity measurements described above (Fig. 5 a). In a higher magnification, both of lighter (Fig. 6 c) and darker laminae (Fig. 6 d) consist of microgranular calcite crystals encrusting dark-colored lines. The lines (recognized better in a lighter lamina ; Fig. 6 c) are 10~30 μm thick and 0.2~1 mm long, and extend mainly perpendicular to lamina boundary. They are probably the traces of cyanobacterial filaments. In the lighter laminae, microgranular calcite occupies the space between the dark-colored lines (Fig. 6 c). Whereas in the darker lamina, the empty space is largely remains (Fig. 6 d). The scanning electric microscopic view displays cylinders of about 10~40 μm in diameter (Fig. 6 e) which seem to have been formed by calcite encrustation around cyanobacterial filaments.

Fluoreluminescence (FL) in a thin section of a tufa provides an undoubted evidence of cyanobacterial association. In FL view of the vertical section, reddish-colored lines can be seen in the uppermost 0.3 mm (Fig. 6 f). The colored lines are dimensionally identical to the autofluorescence of cultured cyanobacteria (Fig. 6 g) which indicates UV-activation of photosynthetic pigments in trichomes. The reddish lines are not seen in the deeper part of specimens where the pigments were probably decomposed (Fig. 6 f). The cultured cyanobacteria is a filament consisting of an unbranched trichome enclosed by sheath. The trichome is stretched chain consisting of uniformly shaped cells, 1~2 μm in diameter (Fig. 6 g). These are the characteristics of family Oscillatriacea.

Discussion

1. Processes controlling the morphological types of tufas

Deposition of tufa is generally controlled by com-

bined effects of chemical, biological, and physical factors. The chemical factors (such as Ca^{2+} content and saturation state) are the basic conditions determining presence and absence of tufa deposits, rather than factors controlling gross morphology. Neither, biological factors (biological association on tufa surface) does not seem to affect the gross morphology. As suggested by Pedley (1990), what determine the gross morphology are rather physical conditions, hydrology and substrate topography.

(1) **Hydrology** : Distribution (Fig. 1 b) and occurrence (Figs. 3 a~e) strongly indicate that the tufas are deposited at the locations covered with strong water flow. This hydrologic condition is irrelevant to all morphological types of the tufas.

The hydrologic control can be recognized even within a tufa deposit. For instance in mound-type tufas, the patterns of lamination shown in Figs. 4 b and 4 c indicate that the depositional rate is larger on the top and the flank of the downstream side than on the overhanging surface. This is due to the presence of strong water flow, which normally covers on the top and the downstream flank (Figs. 3 b and c). The mound-type tufa deposits share similarities in having a tongue-like gross morphology, which protrudes to the downstream and in development of a pool behind a mound (Figs. 2 c and 3 c). These characteristics are reasonably explained by preferential growth to upward and downstream directions.

Correlation between deposition of tufas and strong water flow has been already recognized by Golubic (1969) who described very rapid tufa deposition on the slope of cascades. However, importance of flow strength was best explained by the kinetic models of Buhmann and Dreybrodt (1985) and Dreybrodt and Buhmann (1991), and was later demonstrated in field experiments by Liu et al. (1995). The model of Buhmann and Dreybrodt (1985) indicates that depositional rate increases with flow strength because diffusion of dissolved ions between solid and bulk water flow is enhanced. Depositional rate changes also due to flow condition. According to the calculation of Dreybrodt and Buhmann (1991), depositional rate of calcite in turbulent flow is one order larger than that in laminar flow.

Liu et al. (1995) measured depositional rate of calcite at several locations of different hydrologic settings in

a tufa-deposition stream by measuring weight increase of marble tablets set at the locations. The weight increase means weight of calcite deposited on a tablet. Results of their experiments confirm the kinetic model by showing that the tablets set on slopes of cascade-type deposits recorded much larger weight increase than the tablets in pools.

These models and experiments explain occurrence of the tufas of Shirokawa distributed with strong water flow. In the pools where water slowly flows, calcite precipitates in a slow rate and forms pisoids having only thin calcite cortex (Figs. 3 g and h).

Among the five morphology types, the terraces are formed in a relatively weak water flow. The gross morphology, and also hydrologic circumstances, resembles to rimstone which is normally developed on a spread water passage of weak flow in a limestone cave. The stairs-like morphology is therefore a characteristic associated with the spread passage of weak flow.

(2) **Substrate topography** : Locations of the tufa deposition are the places of strong water flow, such as, steep slopes, waterfalls, and steps formed by boulders. The variation in the proposed types of gross morphology (Fig. 2) basically succeeds to variation in the initial substrate topography. For instance, the cascade type was developed following the previous topography of a waterfall. The substrate of plane slope type is commonly mudstone bedrock forming a relatively smooth slope.

2. Depositional condition of laminated tufas

As mentioned above, our observations indicates that the laminated internal texture shows an annual rhythm of carbonate deposition; lighter laminae developed in summer and darker laminae in winter (Fig. 6 a). Another evidence confirming the annual rhythm was recently presented in the stable oxygen isotopic composition of a laminated tufa in Shirokawa. Matsuoka and Sakai (1998) shows that the $\delta^{18}\text{O}$ changes with lamination; lighter values in the lighter laminae and heavier values in the darker laminae. Because the values of the streamwater are almost constant, the change in $\delta^{18}\text{O}$ of the tufa results from seasonal change in water temperature. Thus, the lighter laminae were deposited in higher temperature and the darker laminae in lower temperature (Matsuoka et al., 1999).

Fig. 6. Microtexture of a laminated tufa at St 9.2 and cultured cyanobacteria. (a) Series of thin sections of the superficial part. Distances in collected location between the six specimens are less than 30 cm. The pictures were taken by using a scanner. (b) A optical microscopic view of the laminated tufa collected in April 1999. The left end is the surface. (c) Microtexture of the uppermost lighter lamina seen in a vertical section of August 1998 specimen. (d) Microtexture of the uppermost darker lamina seen in a vertical section of August 1998 specimen. (e) View of scanning electric microscope of a darker lamina (May 1998 specimen). (f) Floureluminescence (FL) view of the superficial part of August 1998 specimen. The reddish luminescence is autofluorescence of photosynthetic pigment of cyanobacterial trichomes, activated by UV (365 nm). (g) FL view of the cultured cyanobacteria activated by UV (365 nm). (h) Trichomes of the cultured cyanobacteria in a view of differential interference microscope. A trichome appears as a chain consisting of uniformly shaped cells, 1~2 μm in diameter. The cyanobacteria probably belongs to Oscillatoria. Sheaths cannot be seen by this optical method.

As suggested by Yoshimura et al. (1996c), the annual lamination provides chronological constraints as annual rings of wood does. However, further discussion on conditions forming the laminated texture is needed in prior to read the climatic records in the laminated tufas.

(1) **Continuity of deposition** : Regular laminated texture is developed in a place where water covers all through the year. In many cases, the growing tufas of Shirokawa are covered with water only during periods of large water flow (mainly summer and early autumn), and their deposition is discontinuous. Although such tufas may exhibit laminated texture. They consist of accumulation of laminae formed only during rainy seasons, and the boundaries between laminae may represent non-deposition of calcite.

Laminated tufas often display unconformity-like pattern (Fig. 4b). It was probably formed by erosion of the lower set of lamination. Although we have not yet observed the process of erosion, the flood events after heavy rain are most probable.

(2) **Associated biota** : The most important condition in determining the development of the laminated texture is biological. The dominant biota in a depositional site should be cyanobacteria. If the surface is colonized by other organisms, such as mosses and glasses, rhizoids or roots penetrate deep in tufa deposits and disturb layering, as seen in Fig. 4d. Furthermore, organic tissue of mosses is too large, and calcite encrustation on the tissue hardly forms mm-order regular laminae.

The tissue of cyanobacteria consists of delicate filaments, a few μm in diameter and less than $500\mu\text{m}$ in length, and its growth does not inhibit development of laminated structure. The fact that the laminated texture develops under strong water flow, satisfies the hydro-chemical condition for continuous deposition of tufas, and also fits to an independent biological condition, cyanobacteria-dominating biota.

3. Role of cyanobacteria

It has been repeatedly suggested that cyanobacteria biologically plays an important role to induce deposition of tufa (Krumbein, 1979 ; Yoshimura et al., 1996b ; Drysdale and Gillieson, 1997 ; Kohira and Someya, 1997). In the stream of Shirokawa, the depositing tufas are commonly colonized by filamentous cyanobacteria. This fact impresses us that cyanobacteria causes tufa deposition. In this section, we discuss possible effects of presence of filamentous cyanobacteria in deposition of calcite.

Carbonates precipitate in various environments in association with filamentous cyanobacteria. One example is stromatolite which is now depositing in saline water in Western Australia and Bahamas. The cyanobacterial carbonates were also very common in geologic past even to Archean. Because of geological and paleontological importance of such carbonate de-

posits, the role of cyanobacteria has been much discussed in many previous studies and categorized into three by Burne and Moore (1987).

(1) **Trapping and binding** : Cyanobacteria normally has mucous sheath which traps and binds the carbonate sediment suspended by flow and wave actions. This role is applicable to the case of the recent marine stromatolites and lucustrine tufas (Riding, 1979 ; Schneider et al., 1983). In these environments, suspension of the bottom sediment is a common phenomenon.

However, in the tufa-depositing stream of Shirokawa, the water is normally very clear, and sediment suspension is insignificant. Material encrusting the cyanobacterial sheath mainly consists of tiny calcite crystals (Figs. 6c~e). As Kano (1997) suggested, the calcite encrustation is too thick to be regarded as trapped material on sheath. It should be formed by in-situ precipitation. The effect of trapping and binding is probably minor. What the cyanobacterial sheath traps are only quartz and clay minerals.

(2) **Photosynthesis** : Photosynthesis of cyanobacteria can induce calcite precipitation by consumption of dissolved CO_2 species which forms the micro-environment of high pH and increased CO_3^{2-} concentration. This process, categorized in biological-induced mineralization (Lowenstam, 1981), has been reported in several lucustrine carbonate sediments (Taylor, 1975 ; Brune and Moore, 1987). However, it is important in the case of a calm water environment where the water layer of increased pH can be preserved from diffusion. As strength of water flow increases, such high-pH microenvironment is diminished.

The tufas of Shirokawa are deposited mostly under strong water flow. If there is the effect of photosynthesis in case of Shirokawa, it can be important only in a thin water layer covering cyanobacterial filaments. In such a thin and stagnant water layer, dissolved CO_2 can be effectively consumed by photosynthesis, and the resultant calcite deposition does not cause significant decrease of pH in the water layer. In this circumstance, calcite deposition processes with growth of cyanobacteria.

However, this circumstance is inconsistent with production ratio between carbonate carbon and organic carbon of the tufas in Shirokawa. Considering the most effective biological-induced calcite deposition, one mol of CO_2 fixed photosynthesis results in one mol of deposited CaCO_3 (Merz, 1992). Thus, if the deposited CaCO_3 is fully photosynthesis-induced, weight ratio between the organic and carbonate production is 3 : 10 or more (CH_2O is simply adopted for organic matter). Such a high organic/carbonate production ratio is unlikely to produce the textures of the tufas of Shirokawa which consist of thick ($10\sim 40\mu\text{m}$ in diameter) calcite encrustation on thin (less than 5

μm in diameter) cyanobacterial filaments (Figs. 6 c, 6 e, and 6 h). Furthermore, content of organic matter is very minor in the tufa deposits of Shirokawa. Although we did not precisely estimate, organic matter is only a small part of insoluble residue, and the residue constitutes around 10% in weight of the tufas (Table 1). Even considering that organic matter (cyanobacteria) is easily decomposed in the tufa deposits, we assume that the carbonate carbon production much exceeds the organic carbon production, probably in the order of 10~100. Therefore, the effect of photosynthesis is regarded small in the case of Shirokawa.

(3) **Nucleation of calcite crystals** : Cyanobacterial sheath activates crystal nucleation of calcite (e.g., Tazaki and Ishida, 1996). The activation mechanism is associate with molecular structure of the polysaccharide sheath which attracts oppositely charged Ca^{2+} and further nucleation of calcite (Drews and Weckesser, 1982). Once calcite nuclei were formed on cyanobacterial filaments, calcite precipitates in the form of crystal growth.

In terms of kinetics of calcite precipitation (Plummer et al., 1978), precipitation rate is function of reaction surface area. Bushy texture of cyanobacterial filaments is also favorable in activating calcite precipitation by increasing effective reaction surface. These mechanism consistently explain the thick calcite encrustation on the cyanobacterial filaments which is seen in the tufas of Shirokawa (Figs. 6 c~e).

4. Rate of abiotic precipitation and lamination of tufa

The lighter summer laminae are less porous (Fig. 5 a) due to the texture of more densely packed calcite crystals (Fig. 6 c) than the darker winter laminae. As discussed above, the effect of photosynthesis is probably quite small. Thus, development of the laminated texture should be explained by seasonal variation in the condition of abiotic calcite precipitation which related with water chemistry and hydrology.

In the stream of Shirokawa, the water in summer generally records higher Ca^{2+} content, higher temperature, and larger water flow than the water in winter (Kano et al., 1999 b). In a supersaturated freshwater, all of the three physico-chemical condition of summer are positively correlated with the calcite precipitation rate (Plummer et al., 1978 ; Buhmann and Dreybrodt, 1985). Thus, we here propose a hypothesis of development of lamination in terms of seasonal change in the abiotic calcite precipitation rate (Fig. 7). In summer with a large accumulation rate, calcite crystals thickly encrust cyanobacterial filaments, occur even in the free space between filaments, and form a lighter-colored lamina. The upward-growing tendency of the cyanobacteria (e.g. Fig. 6 c) probably results from creeping out of encrustation by calcite crystals. Then once exposed, they become sites for further encrusta-

Fig. 7. Depositional model of a summer lighter lamina (upper) and a winter darker lamina (lower). Larger precipitation rate in summer results in thick calcite encrustation on cyanobacterial filaments and reducing porosity. Whereas in winter, calcite precipitation is limited in close vicinity to the filaments.

tion. Whereas in winter with a small accumulation rate, deposition of calcite is limited near the filaments and forms porous texture of a darker-colored lamina (Fig. 7).

5. Comparison with laminated texture of other localities

Although numerous studies have reported laminated texture of tufas (e.g., Ford and Pedley, 1996), only few studies have carried out repeated sampling and observation for describing 'growth' of lamination and texture change of laminated tufas. Furthermore, as far as we know, none has concluded the layering processes which are similar to those proposed in this paper ; seasonal variation in abiotic precipitation forms the summer lighter (dense) laminae and the winter (porous) laminae (Fig. 7).

Some previous studies described annual laminations which differ in texture from the tufas in Shirokawa. For example, Chafetz et al. (1991) reported that the lamination of the tufas of Oklahoma (U.S. A.) consists of the summer dark (sparitic) laminae and the winter light (micritic) laminae. The material of Oklahoma shows characteristic texture of the summer lamina, exhibiting an aggregate of fan-shaped structures. Each fan, 1~3 mm in diameter, consists of filamentous cyanobacteria in the center fringed by radial-directed

and coarse-grained (0.1~0.5 mm in diameter) calcite crystals (Chafetz et al., 1991). The fan is texturally similar to a shrub which is very commonly seen in travertines of much larger depositional rate (e.g., Sakurai and Kawamura, 1997). Chafetz and Folk (1984) who studied a travertine in Italy, reported that shrubs are normally deposited in summer and forms layering with the winter laminae. In the deposits both of Oklahoma and Italy, the winter laminae are micritic and consist of cyanobacterial filaments and fine-grained calcite crystals; the size of the crystals ranges 40~60 μm in the material of Oklahoma (Chafetz et al., 1991).

The fan- and shrub-shaped structures formed due to a rapid summer precipitation of calcite fringing cyanobacterial filaments. Thus, processes forming the laminated texture of Oklahoma and Italy, as well as the material of Shirokawa, are similarly explained in terms of seasonal variation in abiotic precipitation rate; larger in summer and smaller in winter. Difference in the textural appearance from the tufas of Shirokawa is probably related to water chemistry. According to data represented by Chafetz et al. (1991), the water contains Ca^{2+} in 2.0~2.5 times larger concentration of the water of Shirokawa that causes much higher precipitation rate.

Conclusions

(1) Gross morphologies of the tufa deposits of Shirokawa Town (Ehime Prefecture, southwest Japan) are classified into five; tufa encrusting boulders, plane slope, mound, terraces, and cascade (Fig. 2). The morphological types basically follow the initial substrate topography.

(2) Hydrologic conditions, especially strength of water flow, are important factor controlling tufa deposition and associated biota. Tufas are actively deposited in locations of strong water flow. This relation can be seen even in a single tufa specimen of mound-type morphology (Figs. 4a~c).

(3) Under strong water flow, the surfaces of tufas are colonized by cyanobacteria. Whereas on the surfaces covered with weak water flow, moss is the dominant associated biota.

(4) Laminated internal texture is commonly seen in the tufas of Shirokawa. However, regular lamination tends to be developed in tufa deposits which are covered with permanent strong water flow and are colonized by cyanobacteria. Moss disturbs development of regular lamination (Fig. 4d).

(5) The lamination consists of repetition of dense (average porosity 61%) lighter-colored laminae and porous (average porosity 71%) darker-colored laminae which correspond to summer and winter deposits, respectively. Our observation strongly confirms that the lamination is annual.

(6) For the laminated tufas of Shirokawa, the

annual growth rate mainly ranges from 2.0 to 4.0 mm, and the annual calcite accumulation rate ranges from 0.15 to 0.40 g/cm² (Figs. 5d and 5e).

(7) Microscopically, calcite is deposited as microgranular crystals encrusting filamentous cyanobacteria (Figs. 6c~f). In the lighter-colored laminae, the calcite crystals occur also in the space among the encrusted filaments (Fig. 6c).

(8) The textural difference between the lighter and darker laminae is probably originated from seasonal variation in abiotic calcite precipitation rate (Fig. 7) which is generally large in summer and small in winter. This process also consistently explains lamina-formation of the previously described tufas.

(9) Tufas can be excellent material for climatological study by using annual lamination. Analyses of the stable isotopes (Matsuoka and Sakai, 1998) and minor elements may provide information of terrestrial climate.

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* : in Japanese with English abstract

** : in Japanese without English abstract

〈地名〉

Chikanaga近永 Nakatsugawa中津川 Shirokawa城川

(要 旨)

Kano, A. and Fujii, H., 2000, Origin of the gross morphology and internal texture of tufas of Shirokawa Town, Ehime Prefecture, southwest Japan. *Jour. Geol. Soc. Japan*, **106**, 397-412. (狩野彰宏・藤井秀憲, 2000, 西南日本愛媛県城川町のトゥファ堆積物の形態と内部組織の起源. 地質雑, **106**, 397-412.)

愛媛県城川町のトゥファ堆積物の形態と内部組織は、沢での局所的な水流・地形・生物的条件に支配されている。トゥファは水流の強い場所に堆積し、その形態は5つのタイプに分類され、タイプ間の違いは初生的な基盤地形に関係している。トゥファの内部組織は水流と生物的条件に支配されている。明瞭な縞状組織は絶えず水流に覆われ、表面にシアノバクテリアが繁茂する場所で発達する傾向があり、逆に、断続的な水涸れやコケ類の繁茂は縞状組織の発達を妨げる。トゥファの縞状組織は夏期に堆積する明色で緻密な層と冬期に堆積する暗色で空隙質な層の繰り返しによるものであり、この組織的特徴は水温・Ca濃度・水流に関連した方解石沈殿速度の季節的变化によるものと思われる。縞状組織は“年輪”であり、これを用いることで、古気候解析が可能になるであろう。