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Environmental Factors and Material Characteristics Influencing the Deterioration of the Nikka Stone in the Former Koshien Hotel

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Abstract: Herein, we investigated the physical properties and microenvironments of the tuffs to identify the ways to conserve the two types of tuffs, Nikka and Tatsuyama, utilized in the former Koshien Hotel. We found that the deterioration properties were affected by the swelling of clay minerals in the stones, water transfer within the material, changes in surface temperature due to solar and nocturnal radiation, and differences in wet conditions caused by rainfall. Our findings specifically indicate that the deterioration of the Nikka stone is caused by freezing and that of the Tatsuyama stone is caused by repeated wetting and drying.

1. Introduction

The former Koshien Hotel in Nishinomiya City, Hyogo Prefecture, is a piece of modern architecture designed by Arata Endo and completed in 1930. After extensive renovations to repurpose the building as an educational facility, it is presently used by the Faculty of Architecture at the Mukogawa Women's University. The Nikka stone (from Komatsu, Ishikawa Prefecture), a tuff with a tint of yellow, is an essential aspect of this decorative building and is utilized on the exterior and interior. Currently, the Nikka stone that is used on the exterior is deteriorating in various ways. The Nikka stone is a valuable material that is no longer available because the quarries where it was mined have closed. Consequently, it is vital to determine why the stone is deteriorating and find ways to slow the progress. When the building was renovated, some stones were replaced by the yellow Tatsuyama stone (from Takasago, Hyogo Prefecture); however, this stone has also deteriorated.

These two types of stone are visually similar and difficult to distinguish. However, their defect states are different. Previously, the authors researched the deterioration properties of the stone used for the exterior of the former Koshien Hotel and identified two types of defects, i.e., large deficits and thin detachments. In that study, the authors attributed the difference in the defect states to the location or the microenvironment surrounding the stone (Uno & Noguchi, 2015). Moreover, it was proved that major defects were frequently observed in the Nikka stone and that thin detachment occurred in the Tatsuyama stone (Uno, Iba & Yamada, 2020). These findings suggest that, in addition to the environmental influence, material and hygrothermal properties have a significant effect on the state of deterioration.

Therefore, this study attempts to clarify the difference between the defect states of exterior of the Nikka and Tatsuyama stones with reference to the surrounding microenvironment and the stone's material properties. Therefore, we investigated the microenvironments of the deteriorated stone and the material and hygrothermal properties. Additionally, we estimated the deterioration mechanism for each stone.

2. Material and hygrothermal properties of the Nikka and Tatsuyama stones

The material properties of the samples examined are listed in Table 1, and the items measured in terms of hygrothermal properties are listed in Table 2.

2.1. STONE MATERIAL COMPONENTS

The components of the stone were qualitatively and quantitatively analyzed via X-ray diffraction analysis and thin section examination. Table 3 shows the main minerals that make up each type of stone.

The examination of the thin sections fragments revealed that Nikka stones are primarily formed of mordenite, a volcanic glass substitute for pumice, and contain traces of swellable clay minerals (smectites). The Tatsuyama stone is primarily comprised of quartz and albite, with traces of swellable clay minerals similar to those found in Nikka stones. Swellable clay minerals comprised overlappedplate-like crystals. They expand because of the absorption of water molecules between the plate-like crystals, which contract under dry conditions (Shirozu 2010). This expansion and shrinkage reduces the strength of the stone.

2.2. DENSITY AND PORE SIZE DISTRIBUTION

Images of the outer appearance of the Nikka and Tatsuyama stones are shown in Figure 1, and their physical properties are listed in Table 4. Pore size distribution is plotted in Figure 2.

It is evident from Figure 1 that the Nikka stone is more porous than the Tatsuyama stone. The holes in the Nikka stone are a few millimeters wide, whereas the Tatsuyama stone is quite dense. The dry density of Nikka stones, which contain many voids, is approximately half that of the Tatsuyama stones. In terms of water absorption ratio, which indicates the saturated moisture content of stones, the absorption ratio of the low-density Nikka stone is twelve times greater than that of the Tatsuyama stones.

Table 1 Material properties and measurement methods

Items	Measurement methods
Stone material components	Thin sections of stone were examined with a polarized microscope to confirm the constituent minerals and tissues[1].
Minerals in stone	Clay minerals that cannot be identified using a microscope were quantitatively identified using X-ray diffraction analysis by the indeterminate orientation method and the fixed orientation approach with constant orientation and ethylene glycol EG treatment[1].
Density	Dry densities were measured. The value was obtained after the sample was completely dried at 105°C (Yamada *et. al.*, 2020).
Pore size distribution	The mercury intrusion method was used to determine pore size distribution[2] (JIS-R1655) (Yamada *et. al.*, 2020).

[1] Analysis by Palynosurvey Co., [2] Analysis by Sumika Chemical Analysis Service, Ltd.

Table 2 Thermal and moisture properties and measurement methods

Items	Measurement methods
Thermal conductivity	Thermal conductivity of dry and wet samples was determined using the hot wire technique (ISO 2007).
Water absorption ratio	Water absorption ratio was calculated from the ratio of the water weight in the sample to the air-dry weight of the sample (Yamada *et. al.*, 2021). The water weight in the sample is calculated from the differences between the dry and wet weights. The wet weight is measured after over 750 hours in water. Water absorption ratio is related to the saturated moisture content, i.e., the ratio of the water content in the pores at saturation under atmospheric pressure to the dry weight.
Moisture diffusivity	Moisture diffusivity was calculated from mini disk infiltrometer measurements (Yamada *et.al.*, 2020).

Table 3 Constituent minerals in the Nikka and Tatsuyama stones[1]

	Nikka stone	Tatsuyama stone
Primary constituents	Mordenite	Quarts, albite
Other minerals	Quarts, albite	Orthoclase
Trace minerals	Amphibole, zircon, muscovite, mica-smectite, chlorite-Smectite, etc.	Muscovite, allanite, epidote, titanite, zircon, montmorillonite, etc.

[1] Analysis by Palynosurvey Co.

Table 4 Physical properties of Nikka and Tatsuyama stones (Yamada *et. al.*, 2020)

Physical properties		Nikka stone	Tatsuyama stone
Dry density [kg/m^3]		1180	2260
Thermal conductivity [W/mK]	Dry	0.481	2.03
	Wet	1.14	2.83
Water absorption ratio [%]		39.2	3.40
Void ratio [m^3/m^3] a: derived from the saturated moisture content under atmospheric pressure and dry density. b: derived from the measured cumulative pore volume.		a 0.485 b 0.412	a 0.0844 b 0.147
Saturated moisture diffusivity [cm/s]		2.20×10^{-4}	1.44×10^{-5}

The pore size distribution in the Nikka stone ranges from less than 0.01 μm to more than 100 μm in diameter, with a mode of approximately 0.2 μm. The pore size distribution range of the Tatsuyama stone is ~ ≤0.04 μm, and the maximum value is ~ 0.06–0.07 μm.

2.3. MOISTURE DIFFUSIVITY

The moisture diffusivity is shown in Figure 3, and the saturated moisture diffusivity is given in Table 4.

Under wet conditions, the saturated moisture diffusivity of the Nikka stone is around fifteen times greater than that of the Tatsuyama stone; however, under dry conditions, the opposite is true. From the observations of the dried Tatsuyama stone, the initial stage of water absorption was found to be extremely high. This is due to the large moisture diffusivity of the Tatsuyama stone at low moisture contents.

2.4. THERMAL CONDUCTIVITY

The Nikka and Tatsuyama thermal conductivities are given in Table 4. Under wet and dry conditions, the difference in the thermal conductivity of the Tatsuyama stone is small. However, the thermal conductivity of the Nikka stone is 2.4 times higher when it is wet than when it is dry.

Figure 1 Outer appearance of
the Nikka stone (left) and the Tatsuyama stone (right)

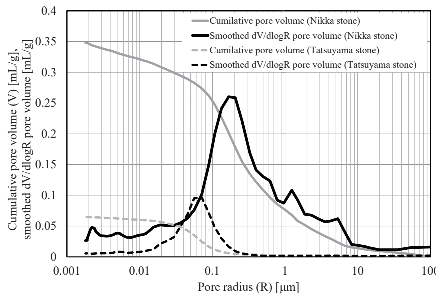

Figure 2 Pore size distribution of the Nikka stone (solid line) and the Tatsuyama stone (dashed line) (Yamada *et. al.*, 2020)

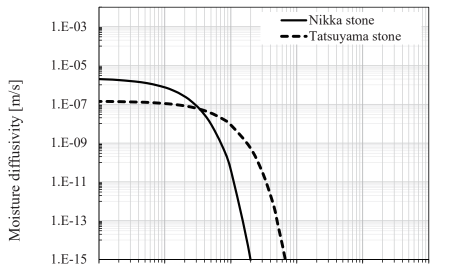

Figure 3 Moisture diffusivity of the Nikka stone (solid line) and the Tatsuyama stone (dashed line) (Yamada *et. al.*, 2020)

3. Defects

Numerous defects were evident in the Nikka stone's horizontal plane, such as the coping stones and stair treads (Figure 4, left). The width of the defects ranged from a few centimeters to a dozen centimeters. The defects were approximately ten millimeters thick. Conversely, with the Tatsuyama stone, the deterioration like the flaking of few-millimeters-thickness on the horizontal and vertical surfaces of the stone beneath the eaves where there was little exposure to rain was observed (Figure 4, right). The deterioration was slight in the area not under the eaves, where the Tatsuyama stone becomes wet in rain.

4. Microenvironment around the stone

4.1. MONITORING SITES

The Nikka and Tatsuyama stones are categorized as the same type of tuff. However, the Nikka and Tatsuyama stones used in the former Koshien Hotel deteriorated differently under similar climatic conditions. To identify the reasons for these differences, we investigated the environmental factors that impacted them.

Figure 4 Defects of the stone materials
(left: Nikka stone, right: Tatsuyama stone)

Figure 5 Nikka stone microenvironment survey (central stairs of rooftop on the third floor); the measured point in the 3rd floor plan (left) and condition (right)

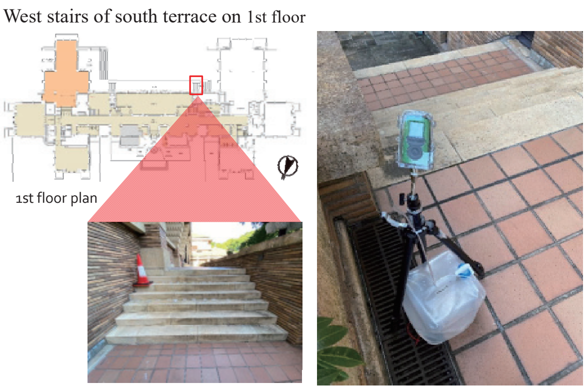

Figure 6 Tatsuyama stone microenvironment survey (south terrace west stairs on the first floor) the measured point in the 1st floor plan (left) and condition (right)

We evaluated the stone's wetness during rainfall, exposure to sunlight on sunny days, and the surface temperature of the deteriorated and sound stones. The outside air temperature was measured on the roof of a separate building near the site. Several measurements were conducted from October 12th 2020 to January 4th 2021. Here, the results between December 29th 2020 and January 1st 2021 are shown.

The third-floor roof terrace was selected as the survey site for the Nikka stone. The measurement point was a north-facing staircase in the center of the terrace (Figure 5). There is no roof at this site, and hence, the horizontal surface of the staircase becomes wet quickly in rain. The stone in the upper part of the staircase has little deterioration; however, previously, cracks have been observed in stones in the lower part of the staircase (Uno & Noguchi, 2015, Isoi, 2021).

The west staircase of the south terrace on the first floor was selected as the survey site for the Tatsuyama stone. The Tatsuyama stone was measured on the west-facing staircase, which has eaves on the north side of the staircase (building side, left section of photo in Figure 6) and a deciduous tree and a low wall on the south side. There is a separate building on the west side. There are evident defects (flaking) under the eaves on the north side of the stairs; however, this rarely occurs at points that are not beneath the eaves.

Figures 7 and 8, respectively, show the air and surface temperatures of the Nikka and Tatsuyama stones in winter.

4.2. NIKKA STONE MEASUREMENT RESULTS

We observed the wet conditions during and after rainfall and found that the water collected under the terrace tile and the tiles flowed to the lower Nikka stone staircase via the back of the upper stair stone. The water from the back side of the stone continued to flow for a few days after it stopped raining.

The surface temperature (Figure 7, left) was lower than the air temperature in both the upper stair (no deterioration) and lower stair (deteriorated) at night. This decline of the surface temperature the stairs is due to nocturnal radiation. During the day, the surface temperature in the upper stair was higher than the air temperature but equal to the air temperature in the lower stair. The differences in surface temperature can be attributed to the exposure to sunlight; the upper stair exposure to the sunlight throughout the day, whereas the exposure of the lower stair to sunlight is only in the afternoon. Here, it is reasonable to assume that, during the day, the temperature in the lower stair (deteriorated)—with less of sunlight and small storage of heat—does not increase much compared to that in the upper stair; thus, at night, the surface temperature of the lower stair also becomes lower than that of the upper stair.

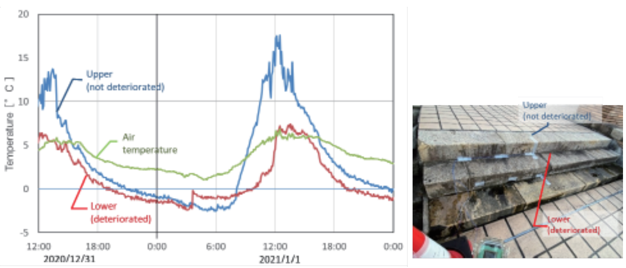

Figure 7 Surface temperature of the deteriorated and not deteriorated Nikka stones (left) and measurement points (right)

From approximately 19:00 to 20:00 on December 31st 2020, (Figure 7 left), the temperature in both the upper and lower stairs was below the freezing point. The lower stair experienced a significant temperature spike of 2°C at 3:40 a.m. on January 1st 2021. This suggests that, after two days of rainfall, the internal water in the stone froze. Observations made a week later determined that cracking had progressed significantly.

4.3. TATSUYAMA STONE MEASUREMENT RESULTS

At the survey site of the Tatsuyama stone, i.e., the stone in the west stairs of the south terrace, during extended rainfall and particularly on windy days, the sound areas (without eaves) became wet, and, subsequently, water flowed under the eaves (deteriorated). Sunlight from the south reached beneath the eaves; however, in sound areas not under the eaves, sunlight is limited by the shadows cast by trees.

According to the measurement results (Figure 8, left), the temperature in the deteriorated area increased on sunny days where the area beneath the eaves was exposed to sunlight. The sound area without eaves was rarely exposed to direct sunlight because it was shaded on the south. Consequently, the temperature did not change significantly. At approximately 12:40 on December 30th, a brief period of precipitation occurred. At the deteriorated area under the eaves, the surface temperature varied by ~6°C before and after the rainfall. It was confirmed that rainfall and sunlight had a significant effect on the surface temperature at the deteriorated site. However, no temperature change was observed in the area without deterioration.

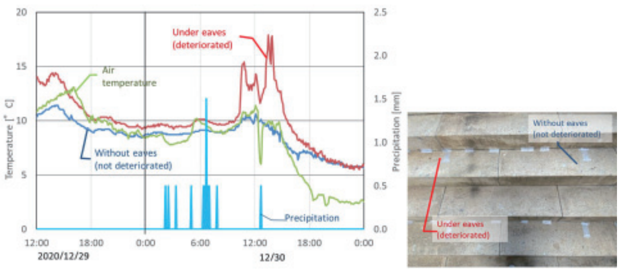

Figure 8 Surface temperature of the deteriorated and not deteriorated Tatsuyama stones (left) and measurement points (right)

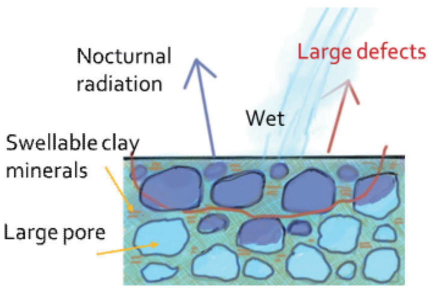

Figure 9 Deterioration mechanism of the Nikka stone

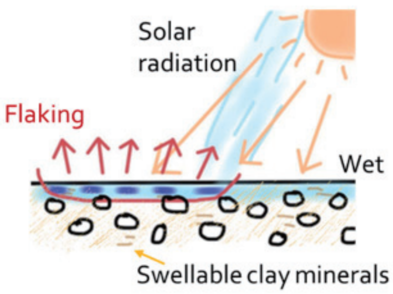

Figure 10 Deterioration mechanism of the Tatsuyama stone

5. Relationship between deterioration and physical properties of stone materials and the microenvironment

5.1. COMMON PROPERTIES OF THE NIKKA AND TATSUYAMA STONES

Both stones contain trace amounts of swellable clay minerals. With repeated water absorption and drying, these swellable clay minerals become brittle. Thus, both stones tend to deteriorate in areas where the moisture content frequently changes.

5.2. DETERIORATION MECHANISM OF THE NIKKA STONE

The Nikka stone includes trace amounts of swellable clay minerals; therefore, frequent water absorption and drying reduces its strength. There are large and small voids in the Nikka stone, and a large defect may emerge if the solid becomes weak in areas with numerous voids. Because the water absorption ratio and saturated moisture diffusivity are large, once water absorption occurs, a significant amount of water diffuses into the deep part of the stone. Since thermal conductivity is not large in the dry condition, the temperature drop inside the stone is small; however, when wet, the temperature drop not only on the surface but also inside the stone. Additionally, ice may form at relatively high temperatures in large-diameter pores, which occur frequently in the Nikka stone.

A diagram of the deterioration mechanism of the Nikka stone is presented in Figure 9. In this investigation, we found that significant deterioration occurred in areas where rainwater was easily absorbed, where the increase in temperature was small during the day, and where the temperature decreased at night due to nocturnal radiation. After a rainfall, the large pores in the Nikka stone contain water. When this water in the pores freezes, it expands and creates pressure, damaging the fragile parenchyma. The large defects are believed to be due to the formation of ice in the internal pores rather than on the surface of the stone.

5.3. DETERIORATION MECHANISM OF THE TATSUYAMA STONE

A diagram of the deterioration mechanism of the Tatsuyama stone is presented in Figure 10. After moisture adsorption, internal absorption is slow. At damaged parts beneath the eaves, the wetness time caused by rainfall is short, and only a thin layer of the surface is wet. When the thin layer is exposed to the sun, the surface temperature of the stone varies significantly, and the surface become dry. The flaking only occurred near the surface because the thin surface layer would experience repeated wetting and drying.

6. Conclusion

To conserve the two types of tuffs utilized in the former Koshien Hotel, we examined the factors affecting the deterioration of the Nikka and Tatsuyama stones by investigating the physical properties and microenvironments of the stone. Our findings suggest that the swelling of clay minerals in the stone, water transfer within the material, surface temperature changes due to solar and nocturnal radiation, and differences in wet conditions caused by rainfall affected the deterioration properties.

In this paper, we investigated the factors and mechanism which caused the stone deterioration, based on the stone material and hygrothermal properties and the measurement results. In order to prove the results, it is necessary to add the reproducibility of the mechanism by the numerical model. In addition, the stone physical properties relating the compression/tension strength should be obtained to show the deterioration of the stone. In the future, we intend to use hygrothermal analysis to obtain a better understanding of the temperature and humidity inside the stone due to boundary condition differences, and the strength of the stone, and to clarify the deterioration mechanism of the stone corresponding to the actual measured results. Additionally, we also intend to explore environmental controls and conservation methods for historic buildings, according to the properties of the stone.

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