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Nondestructive investigation of whitening phenomenon occurred on wood components of historic architecture: in the case of the Confucian Temple of the Kodokan of Mito Domain

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Abstract: The occurrences of the whitening phenomenon of wood in the case of cultural historic architecture have been commonly considered as activities of white rot fungi or degradation caused by solar radiation. Recent studies confirmed that transportation of inorganic compounds with Ca, S and etc. content from stone in vicinity of wood may results in the whitening phenomenon of wood. While in whitening area of wood adjacent to Cu alloy, precipitations of copper or zinc oxalate compounds were confirmed. The whitening phenomenon is observed either in wood adjacent to the stone base or copper components in the case Confucian Temple of the Kodokan (水戸弘道館孔子廟, Mito City, Ibaraki Prefecture). In order to figure out characteristics of the whitening phenomenon occurred in different locations, nondestructive scientific investigation of elemental analysis, microscopic observation, colourimetric measurements were conducted. The investigation confirmed that the whitening phenomenon in wood structures lower than 70 cm height is predominantly affected by Ca, Fe, Cl, S elements, whitening phenomenon of wood adjacent to copper components is predominantly affected by Cu element. Cu element from copper components tends to spread along the longitude direction of the wood. Results of microscopic observation imply that both in “Ca affected” and “Cu affected” wood, the occurrences of whitening phenomenon could be attributed to the formation of precipitation particles among the wood tissues, and degradation of wood tissues.

Keywords: Whitening phenomenon, Wood, Stone base, Copper components, XRF analysis.

1. Introduction

Wooden architecture, vessels, artifacts, crafts and etc. are crucial parts of precious cultural properties of human beings. The durability and values of wooden cultural properties are threatened by degradation of constituent materials or physical damages, which may be induced by environmental factors and human activities in historical time, as well as the worldwide drastic changes of environment in recent decades. Some external factors, such as microorganisms, pests, human activities, natural disasters, sunshine, rain falls, are the well known factors that could induce degradations of constituent materials of cultural properties. As one of degradation phenomenons that generally occurred in the case of wooden cultural properties, the whitening phenomenon of wood could adversely affect the appearance and aesthetic value, and cause worries about the durability.

Generally, the whitening phenomenon of wood occurred in wooden historic architecture would likely be attributed to the external factors white-rot fungi (Blanchette 2000), UV and visible irradiation (Yamamoto et al. 2007). While the concerns about the other possible explanation have

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been raised in recent researches. In the studies of the whitening phenomenon occurred in the wood in contact with base-stones, the occurrence mechanism was described as the accumulation of inorganic components in wood (e.g., Ca, S, etc.), which might be transported by condensation water between wood and stone (佐藤あさひ et al. 2016). On the other hand, the whitening phenomenon of wood is also observed in the area adjacent to metal components in the case of historic architecture, while without contact with earthen or stone base. The previous investigation of this type of whitening phenomenon in the case of the Old Iwasaki-ke Suehiro-bettei Villa (旧岩崎家末廣別邸), confirmed that the existences of metal oxalate compounds, e.g. CuC_2O_4, ZnC_2O_4, etc. might be crucial factors in occurrence of whitening phenomenon (周怡杉 et al. 2016).

These new discoveries implicate that the internal interactions between different constituent materials (stone, metal and wood) of the original structures of historic architecture could cause the whitening phenomenon of wood, which have been neglected in conservation activities so far. In long-term preservation, the unusual phenomenons occurred on components of cultural properties should be treated with cautions and the view of preventative conservation. According to the “minimum intervention” principle of conservation for cultural properties, only the least possible treatments or materials should be undertaken to ensure the durability of the object in conservation activities. The ideal conservation strategies should prevent the adverse development of the whitening phenomenon by controlling external affecting factors, without modification of original structures. For these reasons, it is essential to define different types of the whitening phenomenon. of wood, figure out their occurrence mechanism and affecting factors, and then develop suitable preventive conservation strategies correspond to different types of the whitening phenomenon.

To realize this objective, first of all, profound understandings on the base of scientific case studies about characteristics of different whitening phenomenon are necessary. According to previous studies, wood with different types of whitening phenomenon may present different characteristics of elemental distributions, surface and colourimetric features. These characteristics provide a possibility to recognize different types of the whitening phenomenon via nondestructive scientific methods.

This research attempted to recognize the different whitening phenomenon of wood in the case of historical architecture through nondestructive X-ray fluorescence (XRF) spectroscopy analysis along with colourimetric measurements and microscopic observation. The X-ray fluorescence (XRF) spectroscopy is the most commonly used elemental analysis technique in the studies of cultural properties. The newly developed portable or handheld XRF analysis devices realized the nondestructive in-situ elemental investigation for unmovable cultural properties, since being introduced in the field of art and conservation science from 21th (Parisi et al 2005).

The Confucian Temple is one of the vital constituents of the Kodokan of Mito Domain(特別史跡「旧弘道館」, Mito city, Ibaraki prefecture) which was constructed in 1841, as a school to provide education on Confucianism, history, astronomy, mathematics, music, military strategy, arts, etc., for warriors and their children. The survived site and the Main Gate (正門) including its attached walls(築地塀) was designed to Special Historic Site(1952), the National Important Cultural Properties(国重要文化財,1964) and Japanese Heritage (日本遺産, 2015), as a symbol of educational popularization activities of early modern Japan (畑野経夫 2011; “近世日本の教育遺産群” 2015). The original structures of the Confucian Temple were ruined in 1945. Afterwards, reconstruction was conducted on the survived foundation by zelkova timbers(ケヤキ) in 1970. A partial repair was conducted after the 311 East Japan Earthquake in 2011(Fig.1) (“孔子廟復旧工事” 2012). Nowadays, in areas adjacent to the stone base and copper components of the Confucian Temple, the whitening phenomenon of wood is observed. Although the Confucian Temple is a newly reconstructed architecture, its construction was conducted with traditional materials and well documented. As a constituent of the national historic site,

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contaminations or damages of human activities are mostly avoided for the basic daily managements from related institutions. Based on these reasons, the Confucian Temple was considered as a proper object for this research to reveal the typical characteristics of the different types of the whitening phenomenon of wood in the case historic architecture.

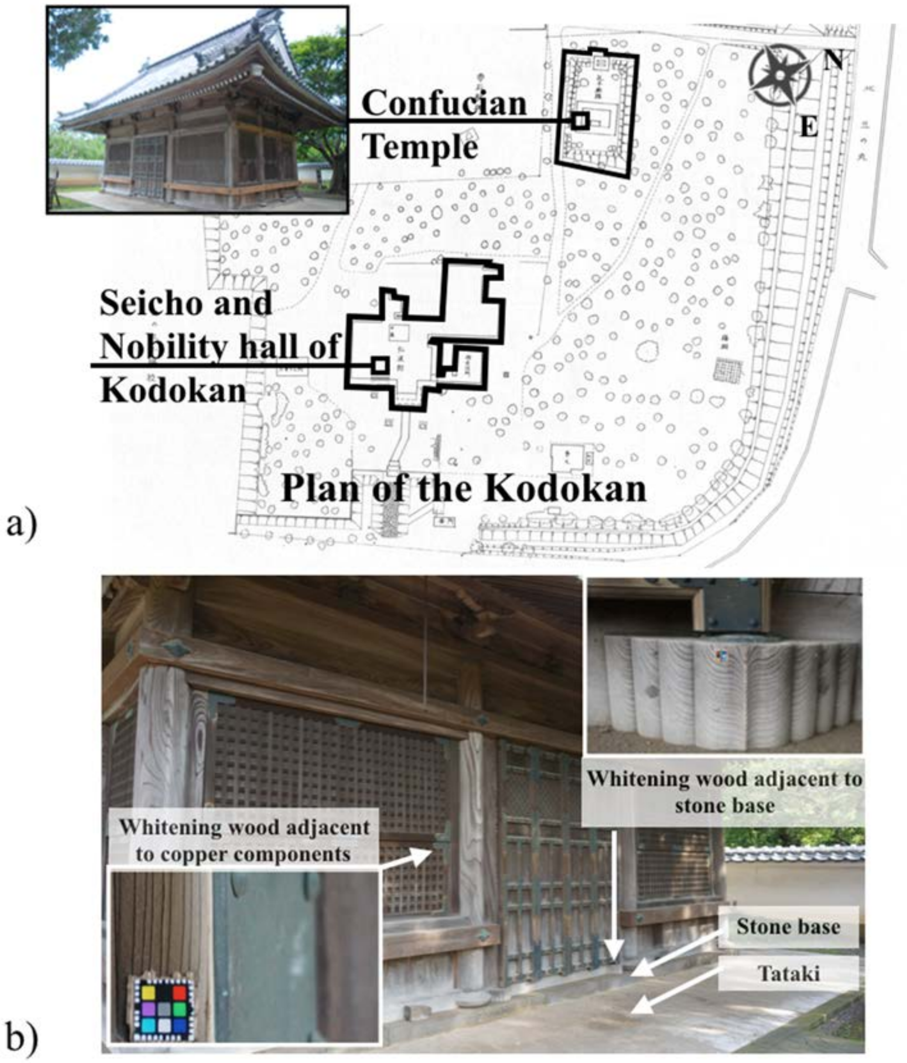

Figure 1. a) Plan of the Kodokan(“弘道館修理工事報告書” 1963) and the photograph of the Confucian Temple(“施設紹介” 2015); b) the 2 different types of whitening phenomenon of wood in the Confucian Temple(Photographs were taken by Yishan ZHOU).

2. Methods

(1) Investigation points

1) Wood structures of the Confucian Temple

The wood structures of the Confucian Temple are separated into 12 sections by the position of its pillars (Fig.2.). The sections were numbered from 1 to 12.

To figure out the elemental distribution in overall exterior sides of the Confucian Temple, investigation points for elemental analysis were determined according to coordinate systems established from 5 to 270 cm height above the bottom (where wood structures in contact with stone base) in the sections with copper decoration components on Nageshi, door and window frames (No. 5, 6, 7, 8, 9, 10, 11 sections); and from 5 to 70 cm height above the bottom in the

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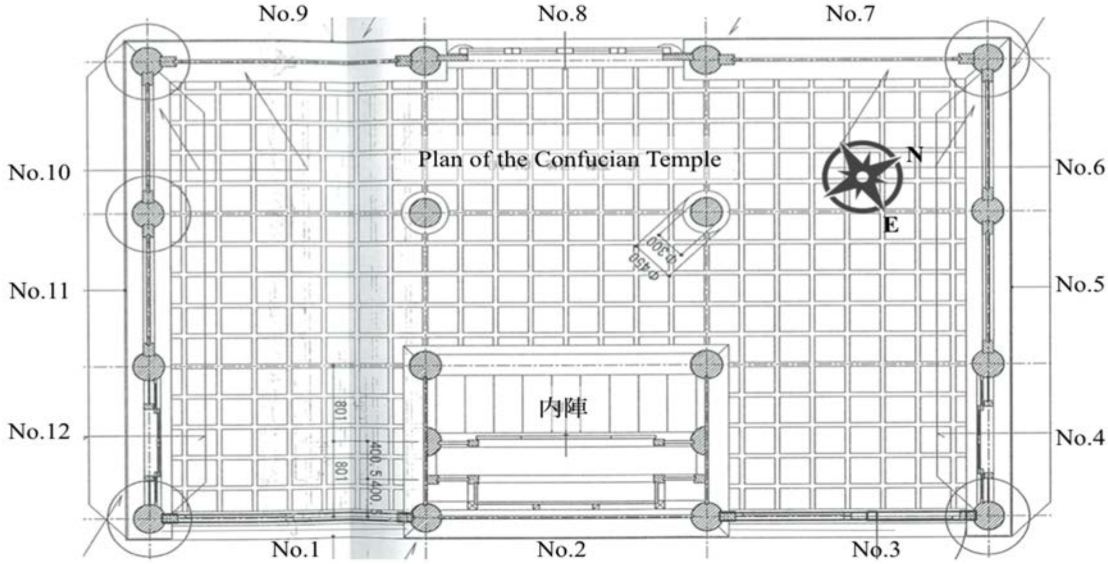

Figure 2. Plan of the Confucian Temple(“孔子廟復旧工事” 2012) and the numbered 12 sections.

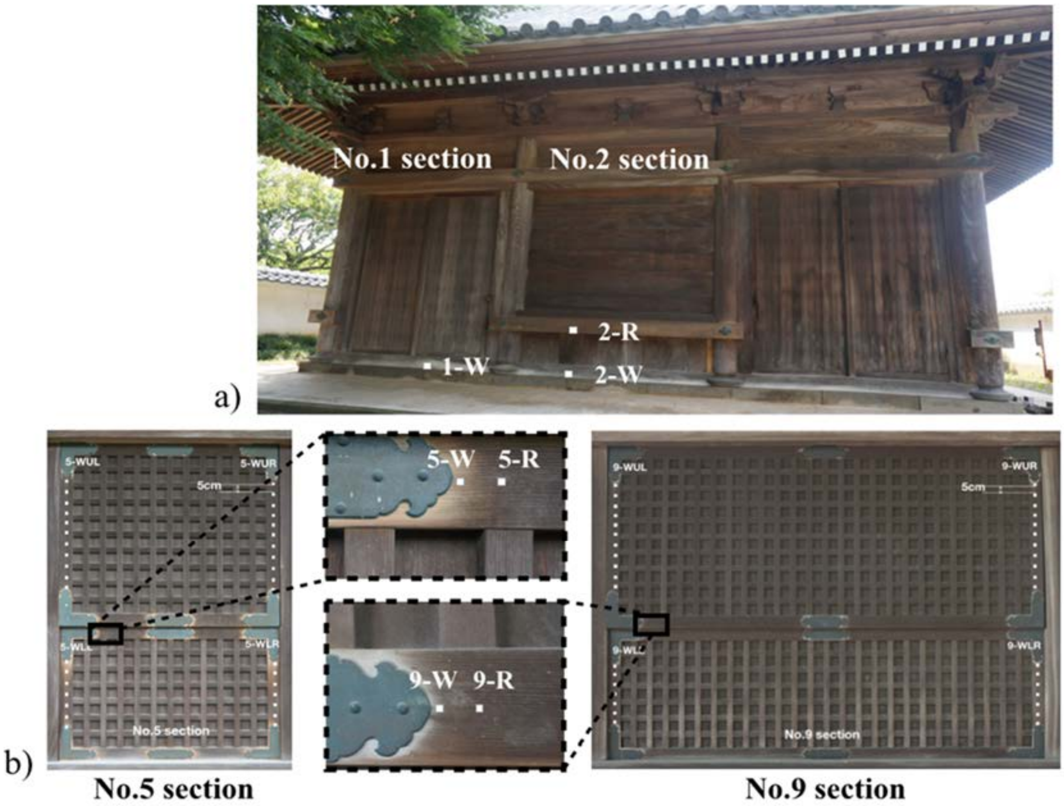

Figure 3. The selected typical points of wood with whitening phenomenon adjacent to, a) stone base; b) copper components and the investigation points of vertical window frames in No. 5 and 9 sections (Photographs were taken by Yishan ZHOU).

sections with almost no copper components (No. 1, 2, 3, 12). Investigation points of wooden structures in all sections were tested generally at 25 cm intervals in the x-axis direction, 5 cm intervals for areas lower than 70 cm height and 10 cm intervals for areas higher than 70 cm height

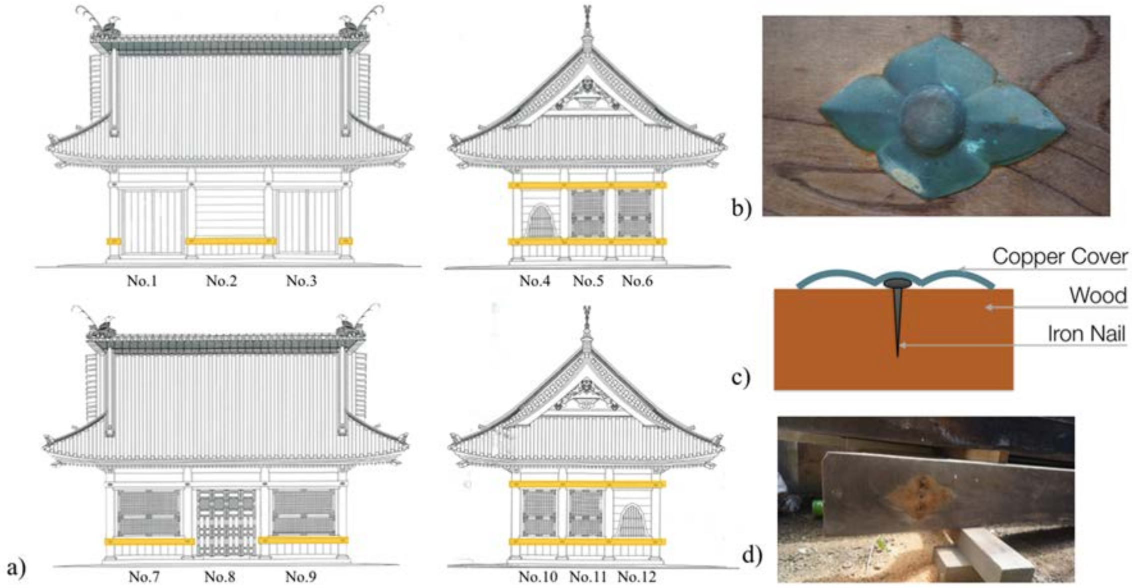

Figure 4. a) The 4 elevations of the Confucian Temple and the original locations of removed Nageshi (orange areas)("孔子廟復旧工事" 2012); b) the type of metallic decorations formerly be installed on Nageshi before removal; c) structure of metallic decoration; d) the investigated removed Nageshi sample in rural environment (Photographs were taken by Yishan ZHOU).

in the y-axis direction. In the Nageshi, wooden parts adjacent to upper or lower edges and 5 cm above or below copper components were tested, and in vertical and horizontal frames of windows and doors, wooden parts between copper components were tested at 5 cm intervals. 4505 points were tested in total. The stone base, the Tataki below the stone base (三和土, earthen floor made from mixtures of lime, water, bittern, etc.), and interior wood of the Confucian Temple were tested as reference points.

The points of wood with whitening phenomenon adjacent to stone base 1-W (in No.1 section, 5 cm height), 2-W (in No.2 section, 5 cm height), whitening wood adjacent to window copper components 5-W(in No.5 section), 9-W(in No.9 section) and reference points 2-R(in No.2 section, 60 cm height), 5-R(in No.5 section, 5 cm left to 5-W), 9-R(in No.9 section, 5 cm left to 9-W) were selected as typical examples to discuss the characteristics of whitening phenomenon at different locations, according to results of elemental investigation, microscopic observation and colourimetric measurements.

The colourimetric measurements for investigation points of vertical window frames in No.5~7, 9~11 sections were conducted to discussing the correlation between Cu content and whiteness degree of wood, with the corresponded results of XRF analysis. The windows were numbered depending on the relative position, for instance, the upper part of window frames in No. 5 section was numbered as 5-WU, and the lower part was 5-WL, and left vertical window frames were numbered as 5-WUL, 5-WLL, the right side were 5-WUR, 5-WLR (Fig.3).

2) Removed Nageshi sample

As shown in Fig.4, copper components are utilized to cover and decorate iron nails that connected pillars and Nageshi (長押, a horizontal piece of timber to connect pillars). Some Nageshi components were substituted in the partial repair of 2011. Since then, the removed Nageshi have been exposed to the rural environment about 4 years, while the parts formerly be covered by the copper components still could be easily distinguished by clearly visible traces of copper components and the less degraded surface.

(2) Investigation methods

The in-situ elemental analysis for exterior sides of wood structures was conducted with a

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portable XRF device Bruker AXS, S1 TURBO equipped with a Silver X-ray tube. The working conditions were 40kV for the potential, 60uA for the current of the tube and 45s acquisition time for each point.

Heights of integrated areas of X-ray fluorescence peaks are related to analyte concentration, matrix elements, and sample thickness, and can be used to reflect quantitive information of analyzed chemical elements (Sitko et al. 2012). In this research, the factor of thickness was neglected, since the relatively large volume of the investigation objects. The detected intensity of each element was applied to represent the relative element content of every investigation point in this research. More detailed elemental distribution surrounding copper components were confirmed by Micro-XRF mapping analysis of one removed Nageshi sample with Bruker M6 Jetstream Micro XRF device, in conditions of Rhodium anode, 50kV for the potential and 600uA for the current of the tube. A (364×180) mm^2 area of the side that formerly be covered by copper components were mapped with 276,480 pixels (500 µm pixel size) and 5ms measure time for each pixel.

The microscopic observation was performed with a Digital Microscope Camera (Nikon ShuttlePix P-400Rv). The colourimetric measurement was performed with a Spectrophotometer (Nippon Denshoku Industries Co., Ltd, NF333), in reflectance measurements mode, under conditions of D65 Illuminant, 10° observer angle and 3-flash averaging mode. The visual differences of whiteness were numerically reflected by the 1976 CIE L*a*b* colourimetric system, in which the whiteness is represented by L* value (+white,-dark) (Johnston-Feller 2001).

3. Results

(1) In-situ XRF analysis for main wood structures

In the case of the stone base and the Tataki, predominant information of Ca and Fe element along with weak information of Cl, Si, K, Mn, Ti elements and etc. were confirmed. In the case of the interior wood point, predominant Ca and K elements along with weak information of Cl, Si, Mn, Fe and etc. were confirmed.

Elements of Ca, Fe, Cl, S, Cu, which are elemental contents of normal wood, stone and copper components in contact with wood structures were focused on in this research. The detected intensities of investigation points were summarized in the form of contour plot graphs with coloured rectangle scales represent intensity levels of elements (growing upwards, from purple to darkish red)(Fig.5.). The lower limit of each element was set as the corresponded detected intensity in the interior wood points, and the upper limit was set as the corresponded the maximum detected intensity in main wood structure points. In the colour scale, detected intensities exceed the lower and upper limit were present as white and grey respectively.

Contour plot graphs of Ca element show significant concentration below 70 cm height. Not only in wood structures, which are directly in contact with the stone base, the significant concentration of Ca were continuously detected until height around 50 cm especially in sections of No. 2, 7(Fig.5.b.). Compared with contour plot graphs and photographs of each section (Fig.5.a.), Ca highly concentrated areas are largely consistent with areas of wood structures with whitening phenomenon.

Beside iron nail affected areas (shown as grey areas in contour plots), the Fe element is mostly significant below 10 cm height, especially in the vicinity of pillar base (Fig.5.c.). In Ca concentrated areas of No. 2, 7 sections, Fe elements present weaker intensities than Ca elements around 50 cm height (showed as blue in contour plots).

Both Cl and S elements were detected in Ca concentrated areas while present different distribution features. Cl element is generally distributed in areas lower than 15 cm height (Fig.5.d.). S element is mainly distributed in areas around 30~50 cm height while not in areas directly in contact with the stone base (Fig.5.e).

Significant concentrations of Cu element were only detected in wood adjacent to copper

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components (Fig.5.f). Contour plot graphs of each section show a similar inverse relationship between the distance from copper components and the I(Cu), especially along the longitude direction of vertical window or door frame wood. Additionally, Cl was also detected in wood in the vicinity of copper components, although not as significant as Cu (shown as blue in contour plot graphs).

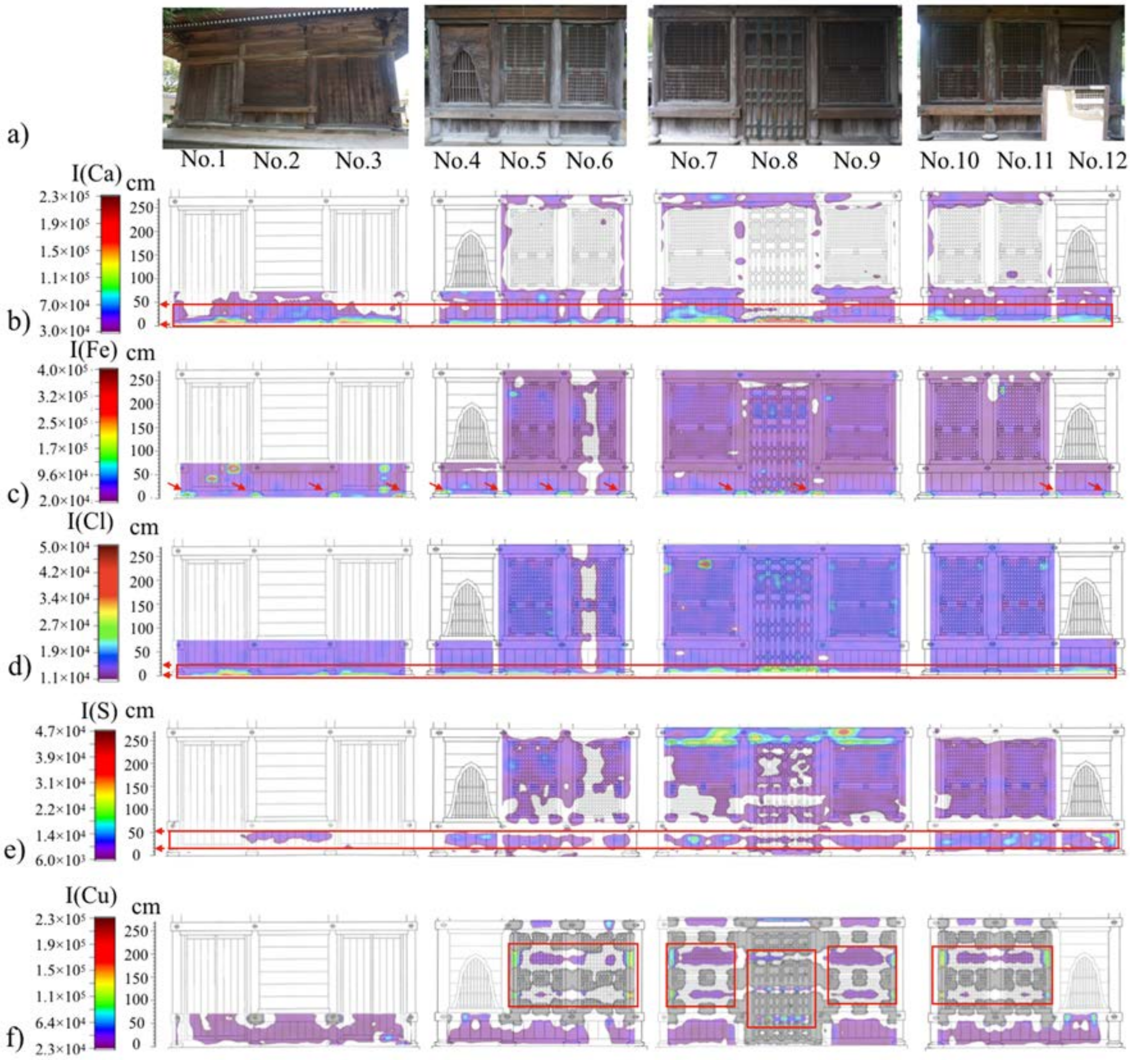

Fig.5. a) Photographs of each section of Confucian Temple; contour plot graphs for different elements, the areas with high element content are presented by red frames or arrows: b) Ca (detected until height around 50 cm), c) Fe element which (mainly detected around pillar base), d) Cl element (detected area lower than 15 cm), e) S element (detected in areas around 30~50 cm height), f) Cu element (mainly detected in wood adjacent to copper components).

(2) In-situ Colourimetric measurements and XRF analysis for window frames

The I(Cu) of vertical window frames of No.5~7, 9~11 sections were graphed versus the corresponded L* value respectively as scatter plots(Fig.6.b.). The scatter plots of No.5, 6, 9, 11 sections seem to present a tendency that whiteness of wood increase with the Cu content. In cases of No. 7 and 10 sections, some points with higher I(Cu) value do not present significant L* value and vice versa. Thus, no discernible patterns are presented in scatter plots of these 2 sections.

(3) Microscopic observation and Colourimetric measurements

The detected intensities of the Cl, Ca, Fe, Cu elements from XRF analysis were graphed as

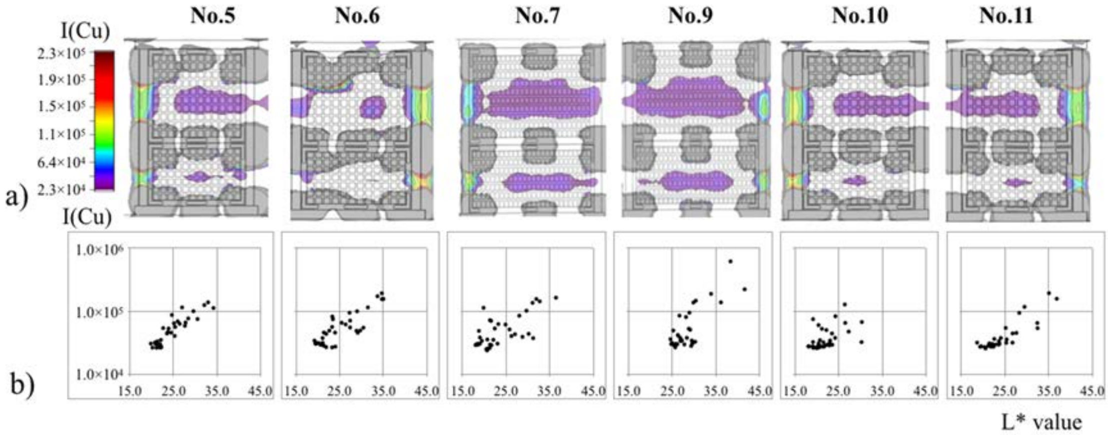

Fig.6 Results of XRF analysis and colorimetric measurements of window frames in No. 5, 6, 7, 9, 10,11 sections: a) the contour plots of I(Cu); b) the scatter plots of L* value(x-axis) versus I(Cu)(y-axis with logarithmic scale).

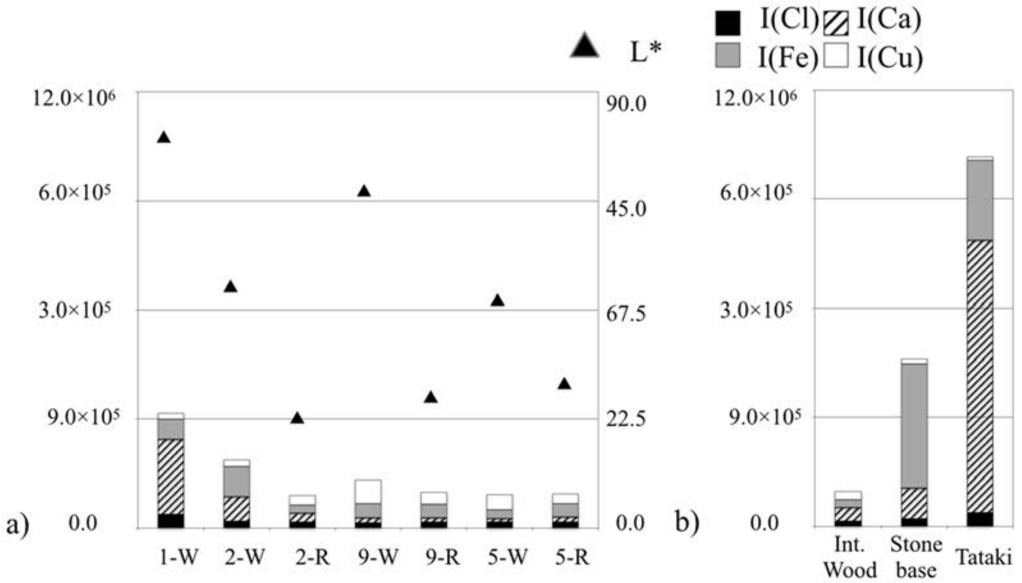

Fig.7 The staked column graph of I(Cl), I(Ca), I(Fe), I(Cu) for: a) points with whitening phenomenon which adjacent to stone base and copper components, with L* values; b) for interior wood point, stone base point, Tataki point.

stacked column graphs with the L* values obtained from colourimetric measurements for the typical points with whitening phenomenon: 1-W, 2-W, 5-W, 9-W points and the reference points: 2-R, 5-R, 9-R points. The graphs are present in Fig.7.a, along with stacked column graphs for interior wood point, stone base point and the Tataki point in Fig.7.b.

All reference points present relatively lower detected total intensities of Cl, Ca, Fe, Cu, and lower L* value around 20~30. The microscopic photo of 2-R, 5-R, 9-R, which are normally degraded wood in the exterior environment, present similar surface characteristics: reddish brown or dark wood tissues with gloss and few attached contaminations (Fig.8).

Microscopic photos of the 2 points adjacent to stone base present different surface characteristics. In the case of 2-W point, which with relatively lower I(Ca) and whiteness degree(L*=49.61), plenty of fine white particles are observed on brown wood tissues. In contrast, in the case of 1-W point which with 3 times higher I(Ca) than 2-W and higher whiteness(L*=80.49), most of the wood tissues are observed as white, while could hardly be figured out if it is due to white attached particles or whitening discoloration of wood tissues.

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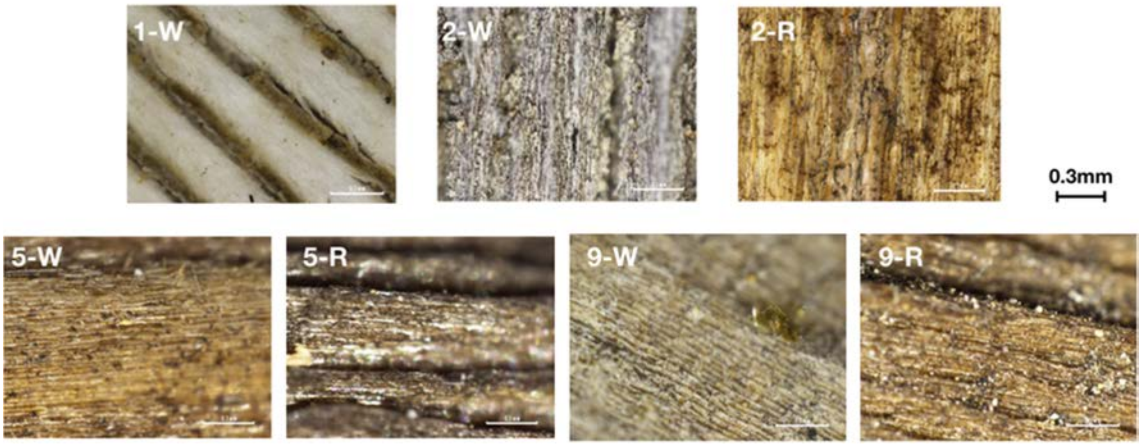

Fig.8 The microscopic photographs of the 1-W, 2-W, 2-R, 5-W, 5-R, 9-W, 9-R points.

Different surface features are also observed in the 2 points adjacent to copper components. The surface feature of the 5-W point, which with relatively lower I(Cu) and whiteness degree (L*=46.81), shows similar characteristics as reference points except for lighter tone of surface colour. However, the surface feature of the 9-W point, which with higher I(Cu) and whiteness degree (L*=69.37), presents as wood tissues with plenty of fine white particles.

(4) Micro XRF mapping for removed Nageshi sample

The Micro XRF maps provide a visualized elemental distribution of the removed Nageshi sample (Fig.9.b.). Significantly different elemental distributions are observed in the case of Cu, Fe, Ca, K elements. The K and Ca elements tend to distribute in the former covered area. Both K and Ca elements are important inorganic content of wood.

The accumulation of Fe element and Cu elements may be attributed to the iron nails and copper component respectively. Fe element is mainly distributed around and below the 2 nail holes along the longitude direction of wood with a downward tendency, in the former covered area. Cu element is distributed around the trace of copper component. Cu element of the upper half side spread horizontally along the longitude direction of wood and downwardly along annual rings into the former covered area. In the lower half side, Cu element trend to spread horizontally along the longitude direction of wood both in outside and inside the former covered area. In the photo of the removed Nageshi sample (Fig.9.a.), a significant whitening phenomenon could be observed in the part that formerly adjacent to copper components, while not in parts distanced from which. The distribution of Cu element in the lower half side seems to be consistent with the whitening areas.

4. Discussion

As the results of in-situ XRF analysis, the characteristic distribution of Ca, Fe, Cl, S elements below 70 cm height may imply a separation process induced by water activities among wood structures, stone base and Tataki for differences of solutes in affinity with stationary and mobile phase（Gaucher 1969）, namely the wood and capillary water. Ca and Fe are 2 predominantly detected elements in the stone base and the Tataki. The capillary water activities may lead to the transportation of elements, which ultimately result in the increase of Ca and Fe contents in wood structures. Although Cl and S elements were barely detected in the stone base, Tataki or interior wood, Cl and S elements could be transported from the surrounding environment to wood via capillary water activities.

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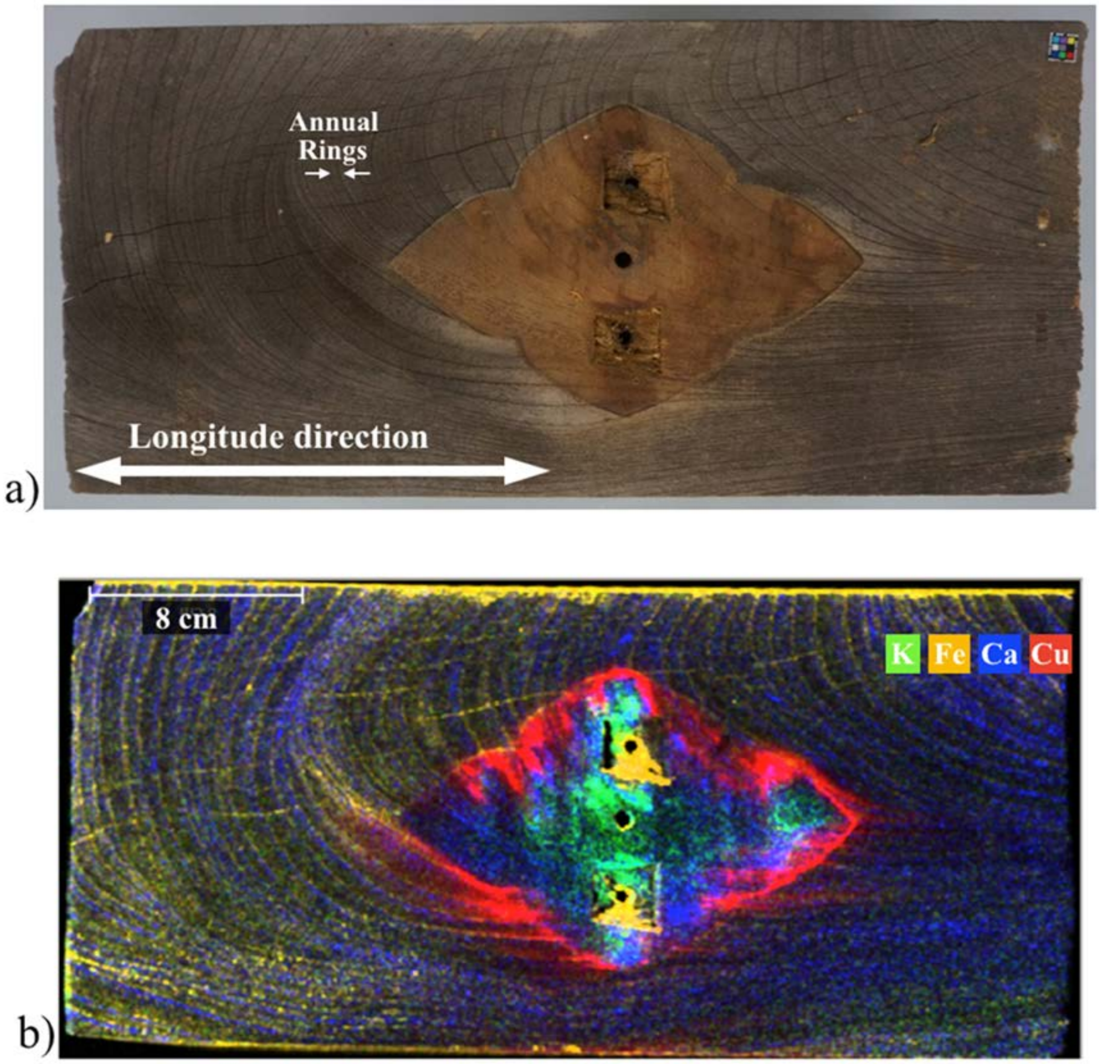

Fig.9 a) The investigated surface of the removed Nageshi sample; b) the Micro XRF maps of K, Fe, Ca, Cu elements (Photograph was taken by Yishan ZHOU)

Microscopic observation of Ca affected wood structure implies that whitening phenomenon in this wood could not only be attributed to Ca-containing white contaminations. The chemical interaction between Ca-containing matters and wood can ultimately result in discolouration of the wood.

In the case of wood in contact with or adjacent to copper components, since metal is cooling medium, relatively high moisture contents are likely to happen around which. This can lead to the transportation of metal elements into the wood (今村浩人 et al. 1987). The visualized image of Cu content for the removed Nageshi sample confirms the Cu element diffusion from the copper component, in the case of a horizontal wood component of exterior side. The copper components may have played a role as shelter for the covered wood. For this reason, the K, Ca content in wood which are readily removed by rainfalls, as well as Fe element from iron nails, tend to concentrated in the formerly covered area. While direct influences of rainfall and gravitational force may have provided a downward influences in the diffusion of copper element at the same time, which can result in the downward tendencies of distributions of the Fe and Cu elements. The copper component may also have sheltered the lower half side of the Nageshi from some direct influences of rainfall. Since the Cu element in the lower half side tend to distributed horizontally along the longitude direction of the wood tissues while not downwardly.

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The investigation of vertical window frames implicates that Cu content in wood and its degree of whiteness could be hardly fit a function of relationship. The microscopic observation of the selected points in window frames may provide the explanation towards this relationship, that Cu affected wood tends to present 2 type of surface features: white particle-attached type, none-particle-attached type. Accordingly, it could be speculated that Cu contents in wood may induce 1) different degradation from normal wood and ultimately result in whitening discolouration of wood tissues, since low valent metal like copper(I) are effective agent in oxidation of organic molecules (Goodell et al. 2003); 2) formation of whitening precipitation particles, such as metal oxalate compounds which have been confirmed in previous studies.

5. Conclusions

The nondestructive investigation for the Confucian Temple of the Kodokan proves that:

- XRF analysis could be an effective method to figure out the elemental characteristics of wood structures with the different types of whitening phenomenon.
- Results of XRF analysis for exterior side present that the whitening phenomenon in wood structures lower than 70 cm height seems to be predominantly affected by elements of Ca, Fe, Cl, S; the whitening phenomenon of wood adjacent to copper components seems to be predominantly affected by element of Cu.
- Micro XRF analysis of removed Nageshi sample confirms Cu element from copper components tends to spread along the longitude direction of the wood.
- A weak linear relationship between Cu content and whiteness degree has been confirmed in the case of vertical window frames.

This study is a preliminary exploration for recognizing the characteristics of the different whitening phenomenon of wood in the case of historic architecture. Through the nondestructive scientific investigation, distributions of specific elements were confirmed respectively in different types of whitening phenomenon. However, microscope observation implicates that both of the 2 types of “Ca affected” and “Cu affected” whitening phenomenon may result from white precipitation particles among the wood tissues, or whitening discolouration of wood tissues.
In the following studies, it is therefore essential to focus on formation mechanism of the white precipitation particles and degradation mechanism of wood tissues along with accumulation process of specific elements such as Ca and transition metal elements in the case of wood historic architecture for 1) grasping the mechanical reduction tendency of wood with whitening phenomenon; 2) figuring out targeted and efficient preventive conservation strategies.

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