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Contribution to the Understanding of Mural Painting Techniques of Jinpari Tomb No. 4 of the Complex of Koguryo Tombs, World Heritage

Hwa Soo Lee and Kyeong Soon Han *

Department of Formative Arts, Konkuk University, Chungju-si 27478, Republic of Korea
* Correspondence: conservation@kku.ac.kr; Tel.: +82-43-840-3677

Abstract: In this study, we have analyzed the chemical components and crystal structures of minerals found in the Jinpari Tomb No. 4 mural paintings, which has allowed us to identify component materials comprising each layer. During the analysis of the painting layer, the green material was identified as malachite. The use of malachite is supported by the high Cu content, the flower-shaped crystals in the microstructures of the painting layer, and the XRD identification results. The ground layer consists of layers of panel-shaped particles. The main component materials were Ca, Al, Si, K, Mg, and Fe, which are usually found in earthen materials. The earthen layer showed high peaks of silicon oxide and calcium carbonate, along with potassium aluminum silicate. The findings indicate quartz, limestone, and mica. The lime layer showed the diffraction patterns corresponding to calcium carbonate, which indicates the use of limestone. The earthen layer consists of aggregated layers of thin panel-shaped structures, with small particles attached around the structures. The lime layer showed aggregations of multi-angle panel-shaped structures and pillar-shaped structures of various types. The analysis has allowed us to shed light on the techniques used in the Jinpari Tomb No. 4 mural paintings.

Keywords: complex of Koguryo tombs; ancient murals; lime; earthen; plastering; painting techniques; conservation

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1. Introduction

Ancient paintings provide vivid insights into the society and culture of ancient kingdoms. They hold great historical, cultural, and artistic values. Mural paintings in Koguryo's ancient tombs were created between the 3rd century and the 7th century for high-class occupiers of the tombs. These paintings are found in Pyongyang, Nampo, and Anak-gun in North Korea. In China, they are found in Ji'an and Huanren. The significance of these murals has gained wider recognition when the "Complex of Koguryo Tombs" was inducted into the UNESCO World Heritage List in 2004. Coupled with China's so-called "Northeast Project," the tombs and murals boosted art history and archaeology projects. Surveys were conducted on the ancient tombs and mural paintings and their conservation status. In 2006 and 2007, South Korea and North Korea joined forced for a joint survey, taking a step further in research efforts to conserve the Koguryo tombs and mural paintings.

The first scientific investigation related to Koguryo mural paintings was the ICCROM report written by Rodolfo Lujan in 1991 [1], which stated that the murals were painted on walls plastered with lime on stonework and the walls were made in three layers with a thickness of 6–7 cm. The first study of Koguryo tomb murals by Korean conservation scientists was an analysis contained in the "Experimental Study on Construction Techniques of Koguryo Mural Paintings" in 1988 [2]. In 2003, the "Study on Painting Techniques of Ancient Murals in the Koguryo Dynasty—Manufacturing Painting Walls" was published. It observed that plastered walls had three layers and clay, sand, chaff, etc. were added to mortar depending on the function of each layer [3]. In 2004, the "Scientific Investigations of

the Tokhung-ri Mural Paintings (408 A.D) of the Koguryo era" was published. The paper was written by Professor Rocco Mazzeo, a pigment analyst from the UNESCO Pyeongyang and Koguryo mural painting survey team, and revealed the coloring technique based on the sample of mural paintings in the Deokheung-ri Mural Tomb [4]. In 2005, the "Study on Pigment Analysis of Koguryo Twin Column Tomb Murals" was conducted [5]. It carried out an in-depth analysis of ground layer and pigment layer using X-ray fluorescence (XRF) and X-ray diffraction (XRD) analysis. From 2004 to 2005, the "Scientific Examination of Mural Paintings of the Koguryo Tombs" was carried out by a UNESCO survey team and the results of the pigment analysis for Deokheung-ri Tomb, Yaksu-ri Tomb, Susan-ri Tomb and Jinpari Tomb No. 1 were published in 2006 [6]. In 2006, a study on the construction technique of Koguryo mural paintings was performed by using mock-up samples made in similar conditions to the murals [7]. Following that study, a study has recently been conducted to understand the characteristics of painting techniques of Koguryo mural paintings [8]. It discovered differences between secco and fresco in terms of the paint film structure of coloring layers based on elemental mapping of mock-up samples created by the two methods. In 2006, North and South Korea conducted a joint survey on material characteristics of eight mural paintings around Pyeongyang [9] and presented the results of the scientific analysis of the walls and pigments based on the samples collected during the survey. In 2007, detailed scientific data on the construction of the lime-plastered walls centering on Jinpari Tombs No. 1 and 4 were found [10]. In 2009, two types of wall samples collected from the Jinpari Tomb No. 4 were examined for microstructure observation, microstructure component analysis, quantitative analysis and mineralogical composition analysis to identify material characteristics of mortar in more detail [11]. Recently, a comprehensive report on the investigation and research conducted by UNESCO for the conservation of Koguryo tomb murals was published [12].

However, scientific research on tomb murals in Korea has been sluggish in recent years, on account of the murals' locations (in China and North Korea), and lack of sustained exchange among researchers. According to the data collected so far, the murals are in poor condition [13,14], and researchers have yet to come up with ways to prevent damage, considering the paintings' materials and other factors.

As such, in this study, we offer information on the characteristics of materials and production techniques for the conservation of Koguryo tomb murals. In particular, we focus on Jinpari Tomb No. 4 (Tomb No. 1 of King Dongmyeong). Mural paintings in Jinpa-ri Tombs hold a great value as they represent changes in the Koguryo culture in the 6th century and their ideas about afterlife, through the transition of the paintings' themes from lotus flowers to *sasindo* (paintings of four guardian gods) [15]. Jinpari Tomb No. 4 is considered in North Korea as the tomb of Ondal and Princess Pyeonggang, and it is also assumed to be the tomb of King Yangwon or the tomb of King Munjamyeong in South Korea. If who was buried in Jinpari Tomb No. 4 is identified, it can serve as the reference point for identifying who were buried in the eight royal tombs of Koguryo built in Pyongyang after King Jangsu moved the capital to Pyongyang in the year of 427 [16].

In particular, the murals at Jinpari Tomb No. 4 are some of the prime examples of Koguryo tomb murals, and studying them will provide us with valuable information on the plastering and painting techniques of Koguryo artisans, as well as other techniques and styles used in Koguryo tomb murals. Specifically, we scientifically analyze and examine the characteristics of minerals comprising the murals, and use the findings to shed light on the techniques used in the murals.

2. Materials and Methods

2.1. Research Site

Jinpari Tomb No. 4 is a corridor-style single-stone chamber tomb that consists of the square-shaped main chamber and a long corridor at the center in the south. The corridor floor was plastered with earth mixed with lime (Figure 1). The walls were built by plastering lime between stones [17]. The murals were painted on the four walls of the main cham-

ber after covering them with plaster. The top half of the walls show paintings of heavenly beings (*cheonindo*), and the bottom half features the four guardian gods. The four guardian gods are depicted with Taoist immortals (*sinseon*), suggesting a stage immediately preceding the typical *sasindo* mural paintings in the late period. The ceiling was decorated with patterns of constellations, lotus flowers, and vines, and ponds were painted on either side of the corridor. The patterns are similar to those on the artifacts excavated from the Tomb of King Muryeong of Baekje, which means they were likely to have been created in or around the early 6th century [18]. Mural paintings in the Jinpari Tombs hold a great value as they represent changes in the Koguryo culture in the 6th century and their ideas about afterlife, through the transition of the paintings' themes from lotus flowers to *sasindo* (paintings of four guardian gods) [15]. In 2006 and 2007, the Inter-Korean Historians Association and the National Research Institute of Cultural Heritage reported that the Jinpari Tomb No. 4 had been damaged beyond recognition [13,14]. While parts of the four guardian gods, the images of the Sun and the Moon, and the heavenly beings are identifiable, it seems difficult to identify the full painting of the murals in their current state.

According to the site investigation report of Jinpari Tomb No. 4 in 2006, the components of the pigments used for each color were estimated by analyzing the major chemical components with a portable XRF. A large amount of mercury (Hg) was detected in the red color on the left side of the north wall, so it was assumed that Cinnabar (HgS) was used, and iron (Fe) was confirmed in the reddish-brown color of the surrounding petal pattern, so it was assumed hematite (Fe_3O_4) was used. A small amount of As was detected in the east wall of the road to the grave, so it was assumed that Orpiment (As_2S_3) was used, and a large amount of copper (Cu) was detected in the green of the tree, so it was assumed that Malachite ($Cu_2(CO_3)(OH)_2$) was used. It was found that gold (Au) was used for the gold color of the west wall and the ceiling of the road to the grave [9,19].

Figure 1. Overview of the Jinpari Tomb No. 4. (**a**) Outside [20]. (**b**) Inside.

2.2. Sample of Mural Paintings

In order to explore the wall construction and painting techniques of the mural paintings in Jinpari Tomb No. 4, we analyzed small amounts of a sample recovered from the corridor floor. The sample consisted of a section sample made with epoxy resin to see the layer structure of the mural paintings, and the sample was designed to understand the characteristics of materials comprising each layer (Figure 2).

2.3. Optical Microscopy (OM)

We observed the surfaces and sections of the sample to understand the layer structure of the mural paintings, their thickness, and the mixture of materials. We used an optical microscope (Axio Zoom V16, Carl Zeiss, Oberkochen, Germany) to observe the sample.

Figure 2. Status of mural sample (**a**) Front, (**b**) Back, (**c**) Side.

2.4. Scanning Electron Microscopy (SEM) with Energy Dispersive Spectroscopy (EDS)

We studied the structure of the sample and the microstructure characteristics and components of the materials. We observed the microstructure of the sample using a normal scanning electron microscope (SEM; SU 3900, HITACHI, Tokyo, Japan), and used the energy dispersive spectroscopy (EDS) attached to the SEM to analyze their chemical compositions. We also used a high-resolution field emission SEM (FE-SEM; JSM-7610F, JEOL, Tokyo, Japan) and an EDS to analyze finer structures. In order to obtain clearer images with higher contrast, we coated the surfaces with Pt and analyzed them using backscattered electron imaging (BSEI).

2.5. X-ray Powder Diffraction (XRD)

We used X-ray powder diffraction (XRD) to analyze the crystal structures of materials comprising the painting layers of the ancient tomb sample and the tomb walls. The conditions for the analysis were: X-ray tube acceleration voltage at 40 kV, electrical current at 30 mA, scanning angle (2θ) at 3~60°, scanning step at 0.02°, and scanning speed at 0.4°/min. Minerals were identified from the X-ray diffraction patterns using the peak machining software (HighScore (Plus)) from Malvern Panalytical.

3. Results

3.1. Construction Status of Mural Paintings

In Jinpari Tomb No. 4, mural paintings are found on the corridor and the main chamber. Natural stones were plastered with lime and earth several times to create the wall surfaces for painting. The colors used on the murals are difficult to identify because the surfaces have been contaminated with red-brown smudges and white salt. However, some observable painting layers show the use of green color, white color, red color, gold color, and others. The structure can be inferred from the partially peeled areas of the mural painting on the western main chamber wall. A lime-presumed layer can be seen on the panel-type stone wall, below a layer of red earth mixed with white powder. The earthen layer is covered by the painting layer (Figure 3).

Figure 3. Status of murals in the Jinpari Tomb No. 4. (**a**) West side of burial chamber. (**b**,**c**) Mural painting details (West side on corridor floor).

3.2. Analysis of Mural Painting Sample

3.2.1. Optical Microscopy (OM)

The surfaces and sections of the sample indicate that the mural paintings in Jinpari Tomb No. 4 are comprised of four layers: the painting layer, the ground layer, and the plaster layer (composed of two layers, the earthen layer and the lime layer) (Figure 4). The painting layer is around 14.2 μm~26.2 μm thick, and contains greenish pigment particles (Figure 5). The surface of the painting layer is covered with red-brown clay materials and white contaminants, which are presumed to be salt (Figure 6a,b). The ground layer is beneath the painting layer. It is a white layer with relatively even thickness ranging between 85.8 μm and 120.4 μm. Beneath the ground layer is a 3.1 to 45.3 μm thick semitransparent layer (Figure 5). Plaster applied on the wall surface is formed by an earthen layer overlapping a lime layer. The earthen layer shows relatively even thickness ranging between 3.29 and 3.34 mm, and contains earth particles and white particles of various sizes. In addition, materials of various lengths and sizes–which are presumed to be fiber and shells–are mixed together (Figure 4). The lime layer at the bottom of the sample mostly consists of aggregated white particles. The layer contains a large amount of material presumed to be fiber, with some other materials, which are presumably charcoal and earth (Figure 6c). The thickness of the lime layer is uneven, ranging from 0.65 to 1.01 mm, as the layer has been removed from the mural painting walls.

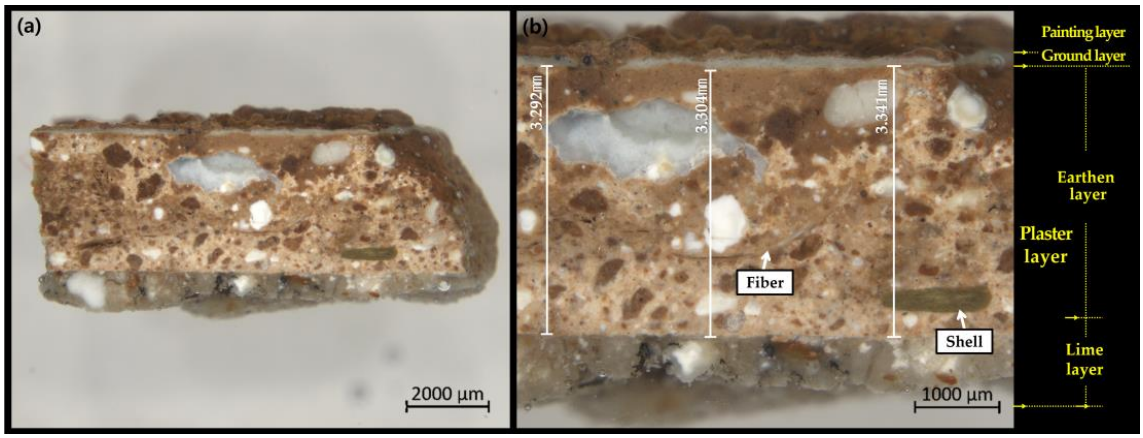

Figure 4. Optical images of cross-sectional mural sample. (**a**) Whole, (**b**) Part.

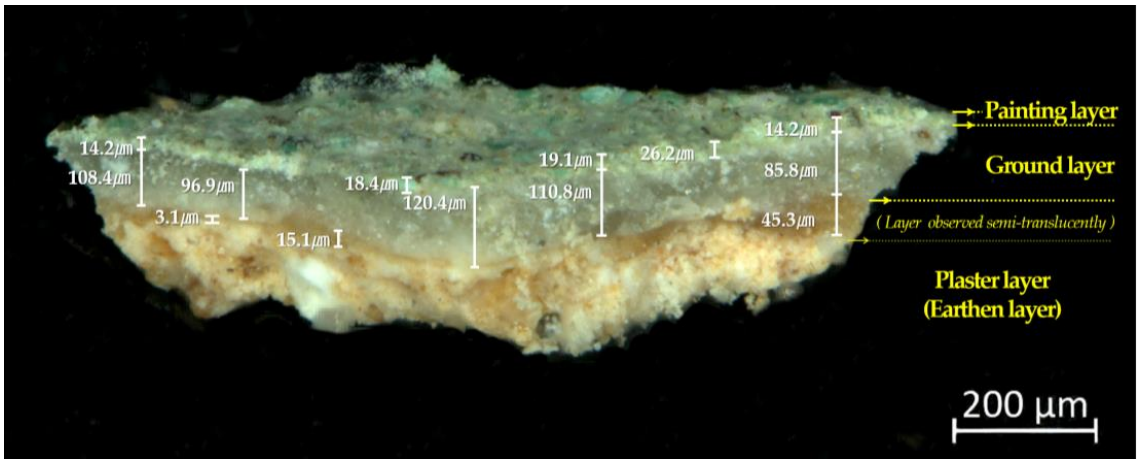

Figure 5. Optical image of a part of mural sample.

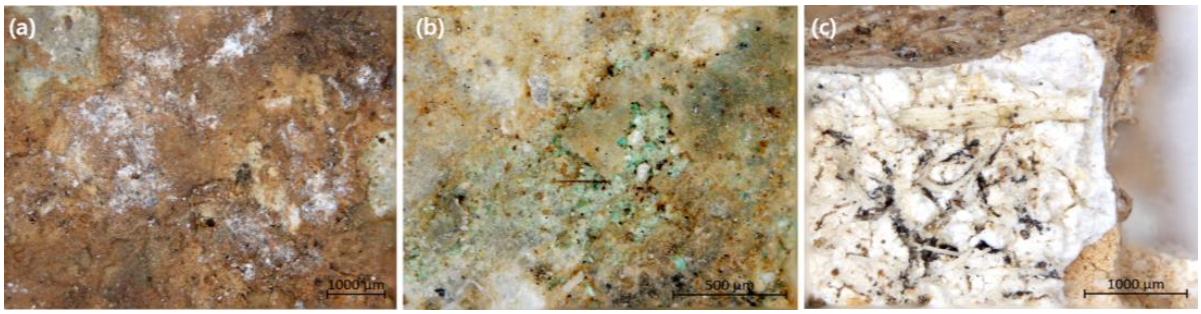

Figure 6. Sample details of mural sample. (**a**) front details: red-brown clay materials and white contaminants, (**b**) front details: status of pigment in painting layer, (**c**) back details: Mixture of lime with materials presumed to be fiber, charcoal, etc.

3.2.2. Scanning Electron Microscopy (SEM) with Energy Dispersive Spectroscopy (EDS)

Then, we observed and analyzed each layer of the Jinpari Tomb No. 4 sample. An analysis of the painting layer and the ground layer showed clear difference in particle shapes between the two layers (Figure 7). The painting layer showed needle-shape structures aggregated in spherical shapes (Figure 7d). The ground layer consists of layers of panel-shaped particles (Figure 7e). A chemical composition analysis of each layer identified the main components of the painting layer as Ca, Cu, K, and Fe, suggesting the use of green pigment containing copper (Figure 8a). The main component materials of the ground layer were Ca, Al, Si, K, Mg, and Fe, which are usually found in earthen materials (Figure 8b). The semitransparent layer beneath the ground layer posed more difficulties in identifying structural characteristics and showed a high Ca content (Figure 8c). Similar characteristics were identified in the sample mapping results (Figure 9). The mapping confirmed Cu as one of the main elements on the topmost layer, with the ground layer mostly consisting of Al, Si, Ca, and K. Ca, which were found across the sample. The Ca content was higher in the semitransparent layer, with lower Al and Si contents. The material compositions were clearly different among the layers.

An analysis of the plaster layer, which consists of the earthen layer and the lime layer, the two component layers were found to consist of particles of different shapes. We analyzed the mixture of materials based on the brightness difference across different atomic numbers on the backscattered electron images of each layer from the section sample. The earthen layer consists of aggregated layers of thin panel-shaped structures, with small particles attached around the structures (Figure 10a). The lime layer showed aggregations of multi-angle panel-shaped structures and pillar-shaped structures of various types (Figure 10b), along with materials presumed to be fiber and shells (Figure 10c,d). A chemical composition analysis of each layer showed that the earthen layer consists of Si, Al, Mg, Fe, Ca, and K, and the lime layer consists of Ca, Si, Mg, and Al (Figure 11). The BSEI of the finishing layer of the section sample indicated particles with different sizes and brightness in the earthen layer, with the lime layer showing brighter particles (Figure 12). The findings indicate that the latter layer consists of a mixture of various lightweight materials. The materials presumed to be fiber in the earthen layer consisted of bundles of thin fibers (Figure 12d). The material presumed to be shells had similar microstructures to those found in other tomb mural walls (Figure 12e).

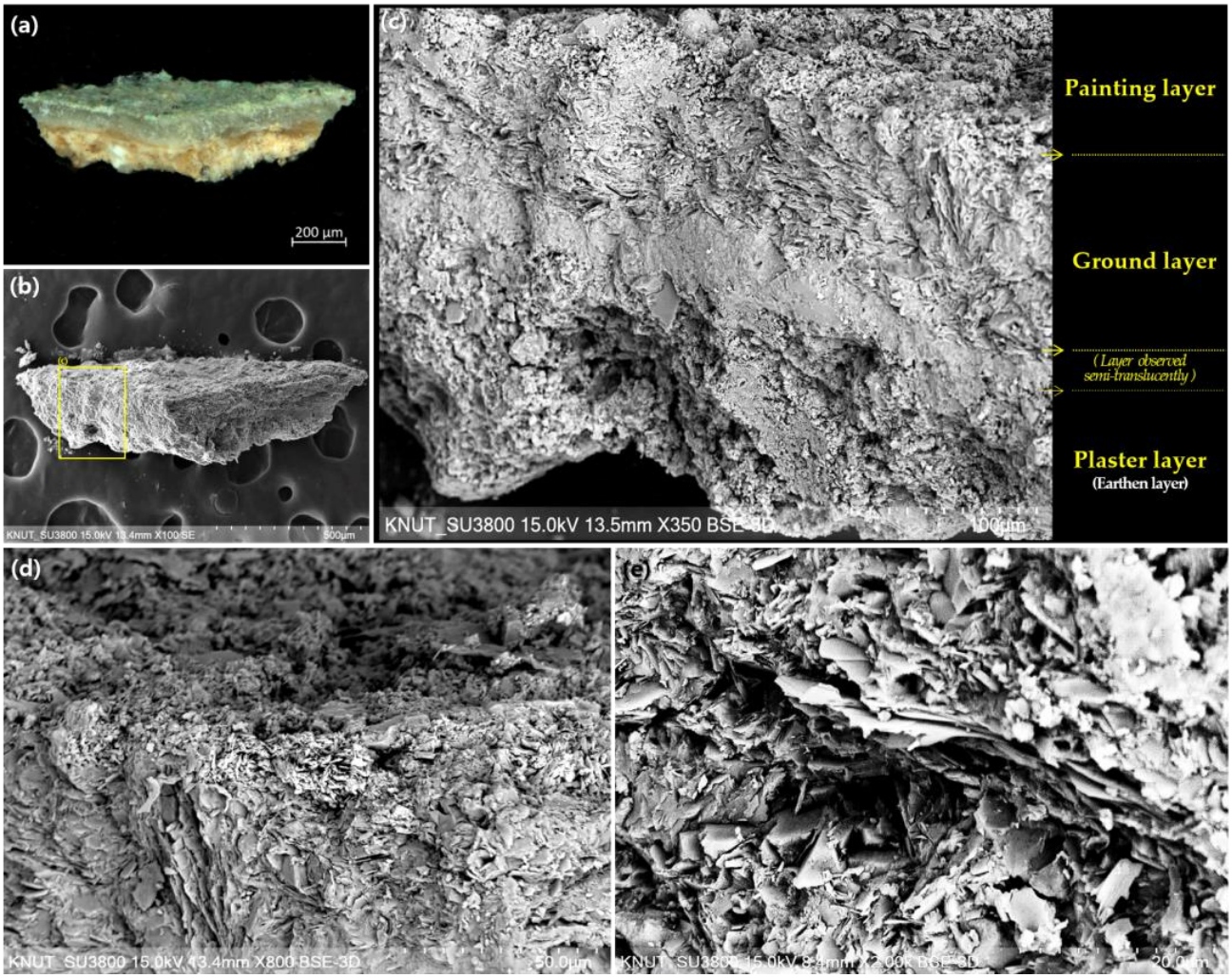

Figure 7. Scanning electron microscope (SEM) image of a part of mural sample. (**a**,**b**) Whole. (**c**) details. (**d**) details of painting layer. (**e**) details of ground layer.

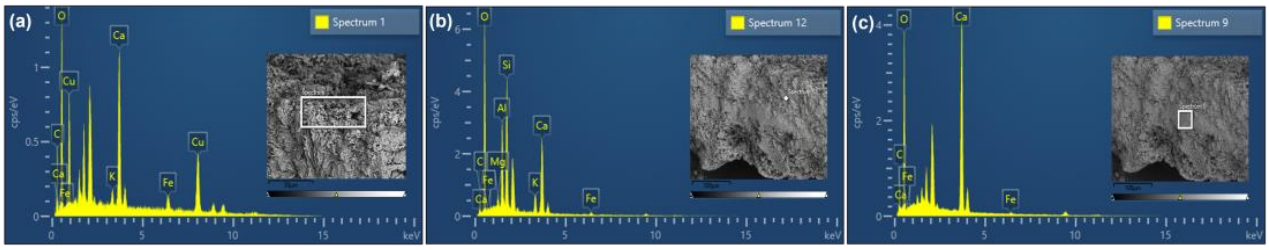

Figure 8. Result of Energy Dispersive Spectroscopy (EDS) of a part of mural sample. (**a**) Painting layer. (**b**) Ground layer. (**c**) The semitransparent layer beneath the ground layer.

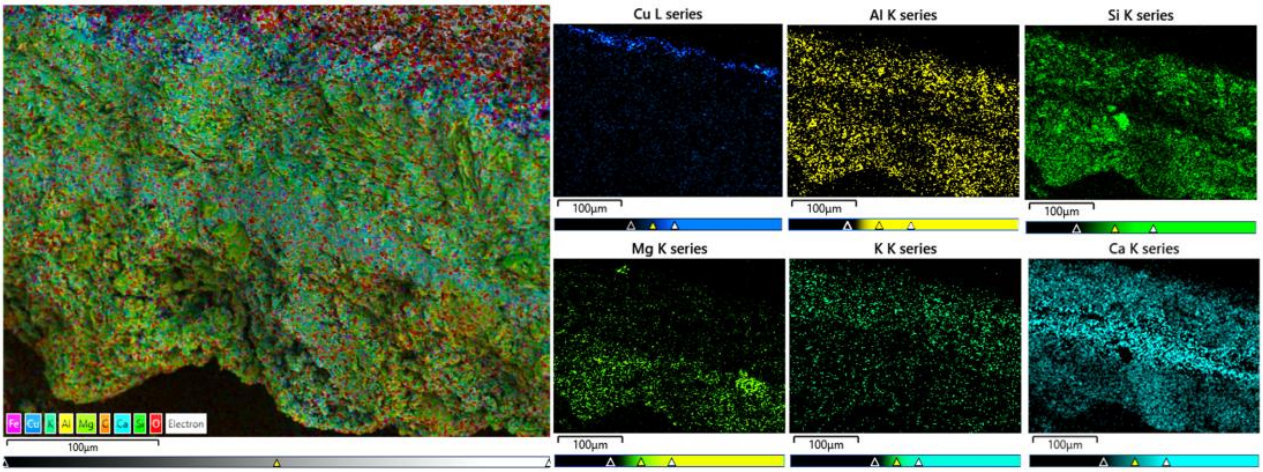

Figure 9. Result of elemental mapping of a part of mural sample.

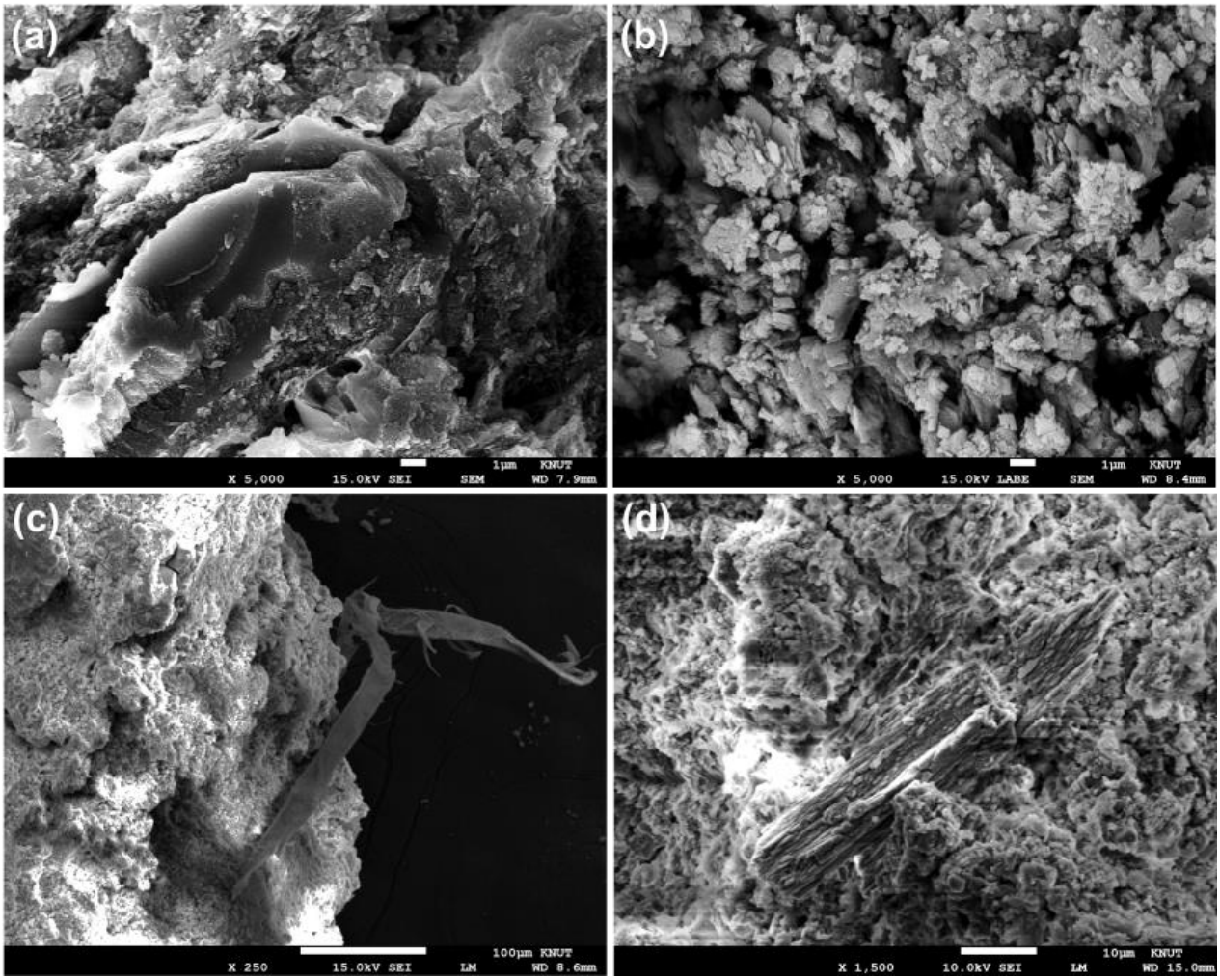

Figure 10. Scanning electron microscope (SEM) image of a part of mural sample. (**a**) Earthen layer. (**b**) Lime layer. (**c**) Fiber in lime layer. (**d**) shell in lime layer.

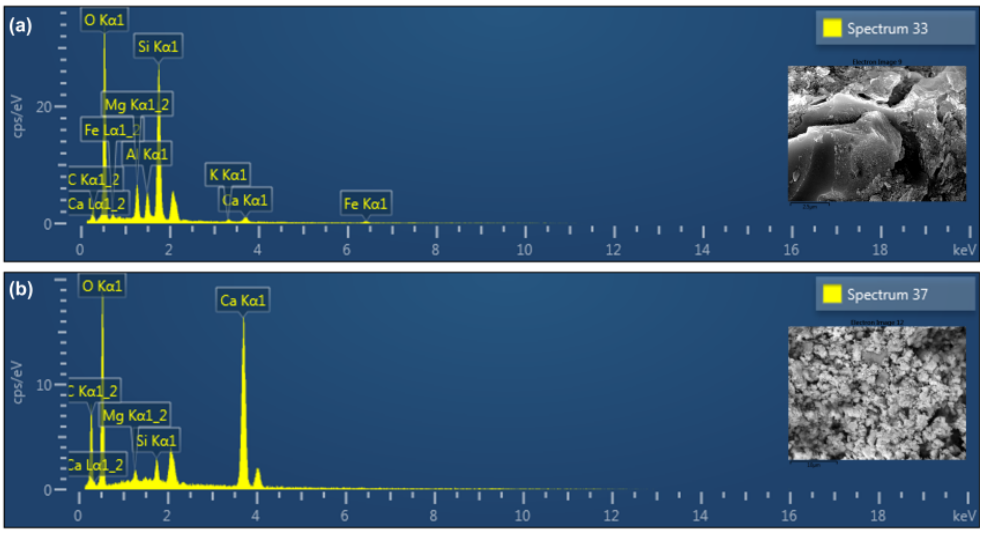

Figure 11. Result of Energy Dispersive Spectroscopy (EDS) of a part of mural sample. (**a**) Earthen layer. (**b**) Lime layer.

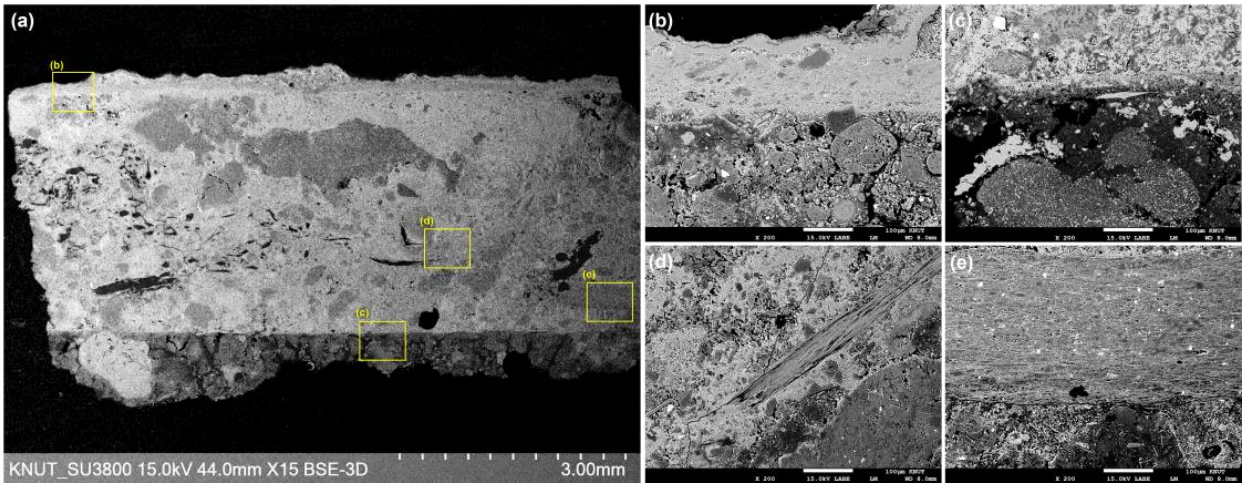

Figure 12. Scanning electron microscope (SEM) image of cross-sectional of mural sample. (**a**) Whole. (**b**) Details: ground layer and earthen layer. (**c**) Details: earthen layer and lime layer. (**d**) Details: Fiber in earthen layer. (**e**) Details: Shell in earthen layer.

3.2.3. X-ray Powder Diffraction (XRD)

We identified the minerals contained in the painting layer, the earthen layer, and the lime layer of the Jinpari Tomb No. 4 mural painting sample, by analyzing their crystal structures (Figure 13). Copper carbonate hydroxide, silicon oxide, and calcium carbonate were detected from the painting layer, which identified the key component material contributing to the green painting as the malachite. The earthen layer showed high peaks of silicon oxide and calcium carbonate, along with potassium aluminum silicate. The findings indicate quartz, limestone, and mica. We also analyzed the white particles in the earthen layer, and identified it as calcium carbonate. Thus, the white particle was confirmed to be limestone. The earthen layer seems to have been created by mixing earth (the main material) with some slaked lime (calcium hydroxide). The lime layer showed the diffraction patterns corresponding to calcium carbonate, which indicates the use of slaked lime. The slight peaks for silicon oxide indicate trace amounts of soil.

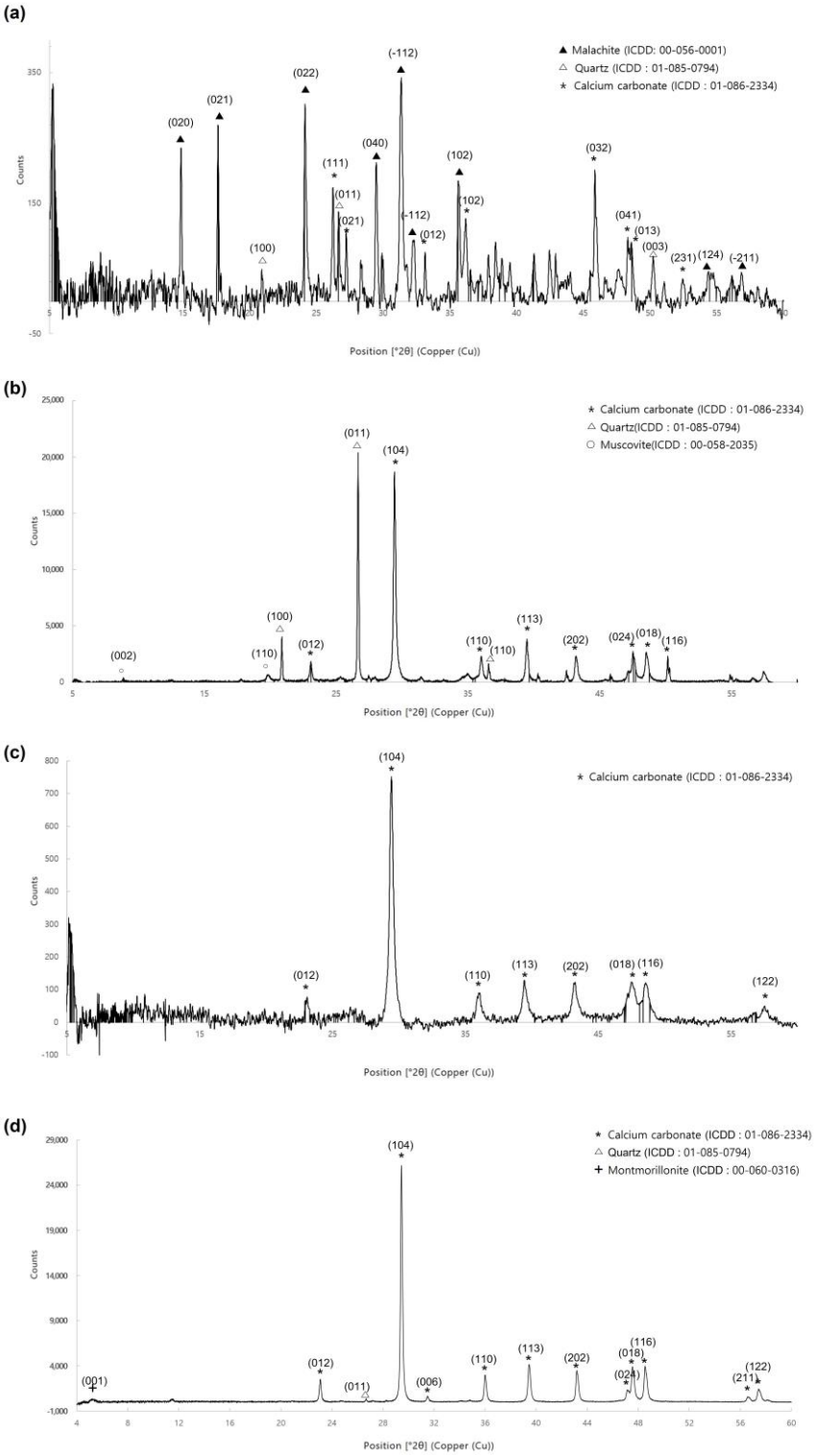

Figure 13. X-ray diffractometer (XRD) spectrum of mural sample. (**a**) Painting layer. (**b**) Earthen layer. (**c**) White particle in earthen layer. (**d**) Lime layer.

4. Discussion

The analysis of the Jinpari Tomb No. 4 sample and layers suggest the following process for creating the mural paintings. Lime plaster was plastered on the surfaces of natural stones or processed panel stones comprising the interior of the ancient tomb, and finished with earthen plaster mixed with lime to create the finishing layer. Then, white earthen materials were used to paint the background, followed by the painting process.

The analysis of the painting layer did not identify the use of various painting pigments. However, the green material was identified as malachite. The use of malachite ($Cu_2CO_3(OH)_2$) is supported by the high Cu content, the flower-shaped crystals in the microstructures of the painting layer, and the XRD identification results. The malachite was widely used to make green pigments for ancient paintings. The findings are consistent with the analysis of green pigments from other Koguryo tomb mural paintings and Gaya tomb mural paintings [14,21–23]. In general, when copper is detected in a chemical analysis, the material is presumed to be copper-type pigments such as malachite or atacamite, or copper rust-type pigments. The contribution of this study includes the clear identification of ancient green pigment through the analysis of diffraction patterns as well as chemical components.

The white earthen layer beneath the painting layer was confirmed as the ground layer. The existence of the layer poses various issues regarding conservation and restoration, as well as the painting techniques used in the mural paintings. The ground layer from our sample was created after plastering the finishing layer. The chemical and crystal structure analysis of the ground layer confirmed white earthen materials in the ground layer. This finding is similar to the white earth-based background painting recently found in the lower part of the painting layer of the Goa-ri tomb murals. This finding contradicts the existing theory that presumed the use of white lead in the background painting for Koguryo tomb murals, which were based on the analysis of murals in the Ssangyeongchong Tomb [5,24]. A study on the mural paintings of the Kitora Tumulus, which is known to have been influenced by the Koguryo mural paintings, found the use of white lead in the ground layer [25]. More in-depth analyses on more murals are required on the issues regarding the relationship between the ground layer and painting techniques. The materials used in ground layers should also be objectively verified using various samples.

The analysis showed different chemical components, microstructures, and mineral crystallizations across different layers. The layer immediately beneath the ground layer had a mixture of earthen materials (quartz and mica, etc.) and lime, which means it constitutes the finishing layer in the mural painting structure. Recent studies reported that the finishing layer of the Koguryo tomb murals was made of high-purity lime plaster [3,7]. However, our findings using crystal structures (XRD), microstructures, and chemical components (EDX and BSEI) indicate the use of earth-based plaster mixed with some lime in the finishing layer of the Jinpari Tomb No. 4 mural paintings. This finding is also inconsistent with the previous findings on the components of finishing layers so far. Our findings create room for a new interpretation regarding the painting techniques used in the Koguryo tomb murals, as they do with regards to the existence of the ground layers. The specific ratio between earth and lime has room for controversy, and additional analyses are required from multiple angles. The calcium layer beneath the finishing layer surface needs to be examined in connection with the crystallization of calcium materials found in the creation process of lime mural paintings.

The lime layer beneath the finishing layer seems to consist of pure lime, and the layer seems to have been plastered on the inner surface of the tombs. The earth and charcoal-like materials found with the microscope seem to have been mixed into the earth and lime during the firing process to make the plaster. The trace amounts of shell-like material found in the lime layer warrants a far more careful analysis. In this study, we were not able to analyze the shell-like materials due to limitations with the sample. However, these materials have been mentioned in previous literature on Jinpari tomb walls [11], and researchers have reported the use of shells in the plaster used on tomb walls from the 5th and 6th-century Baekje and Gaya [26,27]. The existence and identity of the shells constitute significant factors in the study of the conservation of ancient tombs. The walls of Koguryo ancient tombs have been known to be made of processed limestones [2,3,13,14,23]. If a significant amount of shell powder is found in the Jinpari tomb walls, we cannot rule out the possibility that shells were used as lime materials. Only an extremely small amount of shell residue was found in the lime layer of the sample. It means, if shells were indeed

used to create the walls of Jinpari Tomb No. 4, the creators must have had superb firing and processing skills.

Additionally, the plaster layer shows signs of hay and fiber mixed with other materials. The existence of fiber in the walls is connected with the wall's durability. To preserve the mural paintings, therefore, we need more studies on what fibers were mixed with the materials. Even though we were not able to analyze the fibers due to limitations with the sample, further studies are required to compare various cases to identify the fibers and their characteristics.

5. Conclusions

Koguryo's ancient tomb murals reflect the kingdom's class-based society and culture, and contain valuable information on its art, science, and technology. The plaster and painting techniques used in the paintings provide crucial evidence for studying ways to preserve and restore the paintings. In this study, we analyzed the chemical components and crystal structures of minerals found in the Jinpari Tomb No. 4 mural paintings, which allowed us to identify component materials comprising each layer. The paintings were painted on the stone interior surface of the tomb plastered with lime and subsequently covered with earthen. White earth was used on the surface of the finishing layer to form the ground layer, on which the paintings were painted using, among others, green pigment made of malachite. The layers of the paintings could be clearly distinguished based on the characteristics of different materials, which allowed us to shed light on the techniques used in the Jinpari Tomb No. 4 mural paintings.

The sample of this study is too small, and does not contain much information. Therefore, the analysis results cannot reflect all the characteristics of the murals of Jinpari Tomb No. 4. However, the study is a contribution to implement understanding of the techniques used to create the Jinpari Tomb No. 4 mural painting.

A lot is required to preserve mural paintings of the Koguryo tombs. We need to visit more sites and analyze samples for objective verification. However, Koguryo tomb murals are currently inaccessible, and acquiring mural samples is very hard. So, we need to continue with our research efforts using what samples and literature we have available.

One of the consistent findings from scientific analyses on Koguryo's tomb murals is that the murals were created with unique and unparalleled techniques. The plastering and painting techniques of the Koguryo artisans would have changed while affecting, and being affected by, the aristocratic culture and the cultures of the neighboring countries. They would have also affected Koguryo's dealings with the neighboring countries. To substantiate these conclusions, we need further studies on the mutual influence between the techniques of different countries and various tomb murals at the time. Koguryo's diverse tomb mural techniques developed over several centuries, and cannot be reduced to a single style. The academic efforts for mural preservation need to expand into a wider range of disciplines while maintaining awareness on the far-reaching technical scope of ancient paintings.

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