Study on the Conservation and Restoration of Marine Archaeological Porcelains - A Instructive Case of Traditional Crafts from Yue Kiln

Tao Ma[1,2], Xiaodan Liu[1,3], Haiyan Zhu[4], Dan Yang[2*], Zhijun Pan[1]

[1.] Department of Archaeology and Cultural Relics,Nanjing University,Nanjing 210023,China;

[2.] Ningbo museum,Ningbo 315199,China

[3.] School of Arts, Jinzhong University,Jinzhong 030619, China;

[4.] Yinzhou Center of Cultural relics conversation and Management,Ningbo 315199,China

[*]Corresponding author: Dan Yang,1000 Shounan Mid RD,Yizhou

District, Ningbo City, Zhejiang Province, China.e-mail:d.yang@nbmuseum.cn

ABSTRACT：It is generally acknowledged that porcelains, among portable antiquities, boast relatively stable physical and chemical properties．However, marine archaeological porcelains are likely to suffer from deterioration such as salting out, attachment and corrosion due to the complicated burial environments．In the course of conservation and restoration，some natural diseases are easily mistaken for production defects．In order to preserve as much historical information about the antiquities as possible during the clean－up process, we systematically examined traditional porcelain production methods used at the Yue kiln, and conducted further research on typical production flaws that can easily be confused with the natural diseases．Based on these traditional methods and compositional characteristics of porcelains, we provide some guidance on the selection of clean－up methods and repair materials used for porcelains．From the perspective of identifiability and ageing resistance, this paper focuses on discussing the restoration scale of porcelains exhibited in museums．Based on our practical work, we discuss some problems that should be noticed in restoration and also provide an outlook for the future practical work and research．

ADDITIONAL INDEX WORDS: Porcelain; Yue Kiln; Craft; Deterioration; Conservation and restoration; Repairing material

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0 Introduction

Since the 1980s, with the continuous expansion of underwater archaeology, shipwrecks represented by "Huaguang Reef I", "South China Sea I", "Nan'ao I" and "Xiaobai Reef I" and a large number of cargo were salvaged in success ion. Tens of thousands of ancient porcelains also surfaced, placed on the cultural preservation workers' desk. How to deal with such a huge and difficult workload, explore a set of efficient, durable, replicable methods and theories, and put them into practice quickly?

China has a vast sea area, dotted with rivers and

lakes, and a huge stock of underwater cultural relics. Therefore, the conservation of underwater cultural relics has become an important part of China's cultural heritage conservation. On the other hand, all the coastal countries around the world have been seizing the opportunity to develop and utilize marine resources. As a non-renewable resource, the conservation and utilization of underwater cultural heritage have increasingly attracted the attention of governments[1].

For nearly 30 years, with the vigorous development of the underwater archaeological work in China, Ningbo as a port city with a long history, the important anchor of the Maritime Silk Road, the hometown of celadon of Yue kiln which has been the Chinese porcelain since the Middle Tang Dynasty, a large number and variety of China in this shipment are sold overseas. The distribution center of porcelain, which must also contain a large number of celadon from Yue kiln, archaeological discoveries over the years can be confirmed. Therefore, Ningbo must have a large number of potential cultural heritage resources buried in the surrounding waters. Watered-out cultural relics conservation has become an important branch in the field of cultural relics conservation, and the conservation and restoration of watered-out porcelain is one of the key and difficult points[2].

And Yue kiln is a large number of kiln workshop referred in the northeast, Zhejiang province, (today mainly in Ningbo and Shaoxing area), its production techniques appeared in local prehistoric pottery industry directly, is the earliest in the world in the production of mature porcelain kilns, also is one of the technology center of Chinese porcelain, celadon production across the country played a direct or indirect influence, the remarkable position in the history of ceramics.

Starting from the traditional porcelain making technology of Yue kiln, this paper aims to clarify the difference between the defects in firing and the process of use and burial, and make some exploratory studies on the conservation and restoration of this kind of cultural relics, so as to guide the practical work.

1. Defect Identification of Traditional Porcelain Making Process

Ceramics will undergo a series of physical and chemical reactions in the firing process (as shown in the table). In these complex reactions, errors in a certain link will result in various process defects.

Temperature range	Physical and chemical reaction
Low temperature stage (normal temperature ~300℃)	Removal of residual water.
Oxidation decomposition stage (300~950℃)	Exclude structural water, organic matter, carbon and inorganic oxidation, carbonate sulfate decomposition, crystal transformation.
High temperature stage (950~ sintered temperature)	The above oxidation and decomposition continue, produce liquid phase, dissolve solid phase, the formation of new crystal phase and crystal growth, glaze melting.
Glaze melting cooling stage (sintered temperature ~ room temperature)	Liquid phase crystallization, liquid phase supercooling solidification, crystal transformation.

According to the theory of traditional porcelain making technology[34567], the common deformation, kiln crack, glaze bubble, speck, pit, slag dropping, pinprick, orange peel, exposed body, crawling, crazing（惊釉）, smoke staining, body crack, raw firing, overfiring, lamination, inner black, color difference, glaze matte, crystallization and so on

in ancient ceramics can be called defects of traditional porcelain making technology8910. On ancient porcelains, especially on celadon of Yue kiln, the most common defects include kiln crack, shrinkage glaze, slag falling and crazing（惊釉）, and these four defects are also closely related to diseases and conservation and restoration of porcelain cultural relics.

1.1 Kiln Crack

The raw material of celadon used in Yue kiln is mostly "single formula" of china stone, a kind of rock with quartz and sericite as the main minerals, and sericite is a kind of natural white mica, which has a certain viscosity and is similar to kaolin in chemical composition and structure. Sericite has plasticity and also helpful in melting. Therefore, china stone can be used as porcelain material without adding any clay (i.e. single formula). Moreover, high-quality china stone produces a delicate and compact porcelain body due to the low content of Fe_2O_3 and TiO_2. However, a single formula also has its disadvantage. Because of the existence of a large number of glass phase, the thin ware is easy to be deformed in the process of firing. Besides, the temperature control of ancient kiln is not accurate enough, and the temperature is not constant in the firing process, so "kiln crack" is very easy to occur in the groove of the ware (Fig. 1).

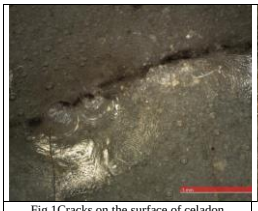


Fig.1Cracks on the surface of celadon

Fig.3 Phenomenon of celadon glazing shrinkage

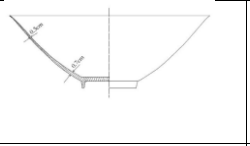


Fig.2Gradually－thickened celadon wall

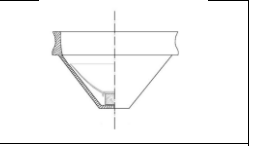


Fig.4Loading and firing mode of celadon placed in a sagger

In addition, in order to adapt to the technique of firing bowl one on top of another (with rings placed as pads inside each bowl), the bottom must support the weight of the body and cannot collapse at high temperature or deformation, craftsmen will make the edge of the mouth thinner and the bottom of the mouth thicker. (Fig. 2), in order to make the wall even, the billet is dried in the dark to a certain extent and then flipped upside down on the fixed mold for tapping. In the process of tapping, while the mold is being organized, some bubbles in the billet will also be discharged due to physical extrusion, thus irregular cracks will often appear on the body, and the biggest factor in this cracking is still the sloppiness of the drilling process.

If this kind of mud is not sufficiently pounded before casting and the air is completely discharged, the bottom will often have a different thickness or even partial stratification after the molding body is formed. and this kind of defect will not only lead to crawling on the glaze, but also produce body cracking. Of course, besides air in the mold, the different water content of each part in the mold is also an important inducement of kiln crack. This cracking may not have penetrated the body, so it did not damage the entire body to produce further crackles. In this case, the cracked part of the body will be filled and covered with molten glaze during the firing process, so that there are no visible cracks in the porcelain after it is sintered, but if the amount of glaze applied does not reach enough to fill the cracks of the body, excessive glaze water will be absorbed in the dry crack, which will make the glaze surface of the implements have a distinct sag feeling, forming defects.

This kind of kiln crack is not the crack caused by external factors, so it should be identified and retained in the conservation and repair process, instead of covering the crack. However, it must be noted that the cracked part on the porcelain cultural relics will be more vulnerable to the erosion of seawater salinity and pollutants due to the incomplete or thinner glaze layer, forming further complicated diseases.

1.2 Shrinkage Glaze

The products of Yue kiln mostly adopt the way that the inner wall swing glaze and the outer wall dip glaze combine to carry on the whole body apply a glaze. Before glazing, the billet is often watered to remove organic matters, dust and other attachments on the billet. However, if it is not cleaned, in the process of glazing, the relatively thick glaze layer of the ware will shrink.

Moreover, the clay material used in the molding will pass through the stale (the mixing of the clay material is not uniform or the humidity is not consistent, which will cause the deformation of the clay due to the uneven shrinkage in the drying and firing process. In addition, the existence of bubbles in the clay will cause the mud body lamination and cracking, in order to prevent these defects, the clay needs to be stale for a period of time in a special stale pool to make uniform moisture, and under the influence of bacteria, organic matter decay of organic acids promote bubble row out can improve the plasticity of the clay.)and mud beating(after the stale clay will go through the combination of manual barefoot and trowel to carry out regular and rhythmic tramping, padding and beating process to promote the uniform clay and the discharge of bubbles, so as to make the clay more uniform and dense.) Nonetheless, if the stale or the beating period is not enough, there will be a lot of undischarged bubbles in the clay. Using this material in the firing process, along with the temperature increases, the water vapor heat expansion, will continue to exhaust, this exhausting process or due to high temperature glaze melt absorbed by stomatal glaze "pinprick" flaw, the phenomenon occurs mostly in the thin glaze layer, such as mouth edge; Or it may cause "bulge" or shrinkage of glaze due to excessive viscosity at high temperature and poor fluidity, which hinders bubble discharge (Fig. 3). Through the observation of vast celadon in Yue kiln, it is found that the gap of the shrinkage glaze is not only unglazed but also stratified in the celadon body. This defect should be distinguished from "damaged glaze" in cultural relic diseases. Although it is not a disease of cultural relics, it has become an inducement to form defects. If the pollutants are continuously invaded in the shrinkage glaze, it will become an active disease. In particular, cultural relics from the sea, where a mass of salt was repeatedly delirized and crystallized, would lead to deglazing in the long run. Hence in the process of conservation and restoration, according to the principle of minimum intervention, it is no need to be filled, but must be thoroughly washed and desalinated.

1.3 slag falling

In the Tang and Song dynasties, many celadons of Yue kiln would adopt one-piece sagger firing mode (Fig. 4) in order to pursue the best varieties. Different degrees of firing adhesion were easy to occur on the edge of the pad and bottom foot. Therefore, a ring of sticky slag would often appear on the bottom foot of the ware. In order to place the mat inside the ring foot, it is necessary to turn the glazed ware upside down to remove the glaze on the inside of the bottom foot, so as to prevent the glaze layer from sintering with the mat while leveling the bottom foot, which is the so-called "mud closing（闭泥）" in the traditional porcelain making process 11 .And if the implementation of the "mud closing" process is not adequately handled, two production defects are likely to occur: one, rotary cutting area and the area of the pad do not overlap, in the case of the rotary cut area is relatively small, the spare part will bedder and glazing part of rotary cut adhesive, there will be a sticky residue after firing, namely a kind of "slag". This slag material is from the pad, and the material of the pad is usually coarse mud with a high content of iron oxide, so there are often tawny and black-brown attachments or "erosion" in the place where the porcelain and the pad adhere (Fig. 5). In addition, the removal of the pad will cause damage to the porcelain body due to mechanical force, which is

similar to the "defect" phenomenon in cultural relic diseases. Second, in the process of rotary cut, the bottom should be filled with water to prevent crack caused by mechanical force. Rotary cut at the same time also can produce viscous because wet mud and adhesive on the artifacts, in the case of not even clear, then there will be a "sticky mud" (in implements a foot mud closed rotary cut down in the process of sludge will without clear adhesive on the device at the bottom of the foot, implements in artifacts appear on the bottom of the foot after firing clastic attachments.) (Fig. 6). Especially in the process of mass production, due to the acceleration of the production speed is more prone to be uncleaned. This is not only the proof of the "mud closing" process adopted by the Yue kiln, but also the appearance of production defects is a kind of basis for speculating the production situation at that time. This kind of defect on the bottom foot is exactly a major feature of traditional porcelain making technology, and it does not affect the integrity and beauty of the ware. We generally do nothing intervention in the process of conservation and repair.

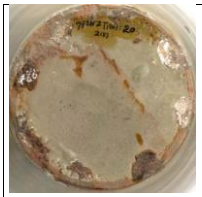


Fig.5 Fireclay cake sticking marks at the bottom

Fig.6glaze slags on the surface of celadon

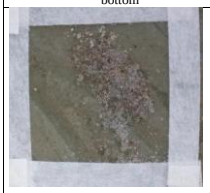


Fig.8 Salt crystallization on the surface of celadon

Fig.7Phenomenon of fissured glaze on the surface of celadon

1.4 crazing（惊釉）

In the long-term storage of Marine environment, under the pressure and erosion of seawater, porcelain will produce simple glaze cracks that have not penetrated the body, which is called "shock crackle（惊纹）". And after firing, there will be a natural cooling process, especially in quartz crystal transformation stage (573 degrees Celsius degrees or so), if cooling speed is too fast, the internal stress cannot be released, there will be a crack on the glaze, namely "crazing（惊釉）", also some people call it "wind crack（风裂纹）" (Fig. 7). This phenomenon is similar to "shock crackle（惊纹）", which is difficult to distinguish. Whether it is in the process of porcelain glaze, or in the later use, preservation process in the process of shock lines are induced other diseases of hidden trouble, seawater salt and pollutants into the channel, so it should be clean up in time, to avoid new diseases caused by environmental changes after the porcelain is discharged from the sea.

2.The relationship between the production, use and burial traces of cultural relics and cultural relics diseases

Respect historical information and the authenticity of cultural relics. First of all, we should distinguish the production, use and burial trace of cultural relics in the long historical process. Among them, burial traces occur in the process of burial and excavation. Due to the physical, chemical and biological effects at various stages, corrosion will occur to different degrees. The damage of these cultural relics is called disease in the work of cultural relics conservation.

According to the degree of stability, cultural heritage diseases can be divided into active diseases, induced diseases and stable diseases. Some production defects and use marks may also cause similar damage, and they will also be considered as diseases of cultural relics when they affect the material survival and integrity of cultural relics.

It is the basis of the principle of conservation and restoration to identify the disease and production traces that need to be dealt with. Due to the limitation of raw materials and technology, there are almost no perfect ceramic products in ancient times. In terms of the conservation and restoration of porcelain, the most urgent task is to

identify the production defects and diseases caused by the use and burial of any porcelain cultural relics, so as to avoid excessive processing of the relics and historical information.

3. Guiding significance of traditional porcelain making technology for conservation and restoration

3.1 Guidance on cleaning methods

According to the analysis of the Yue kiln porcelain making process and the long-term effects of the complex marine environment. Besides a lot of visible stain erosion and induration covering on the surface, there are many soluble salt ions in the body glaze, such as Cl^-, SO_4^{2-}, K^+, Na^+, Ca^{2+}, Mg^{2+}, and the content is higher, basically in the range of hundreds of PPM to thousands of PPM. Affected by the fluctuation of air humidity, these soluble salts will continuously produce crystal evolution (Figure 8) and delight, which will seriously threaten the safety and stability of aquatic porcelain relics. Therefore, the cleaning mainly includes two parts: the first is to remove the stains and induration on the surface of the porcelain; Then, remove the soluble salt ions in the relics. The following aspects should be paid attention to in cleaning and desalting.

3.1.1 Production marks with stable conservation and characteristics of porcelain kiln:

The color of body cracking, slag falling and slime（粘泥）, especially the color of the pad is similar to that of iron oxide, which is easy to be confused with iron erosion and attachment should be retained in the process of cleaning and desalting.

3.1.2 Manufacture of defects induced diseases:

Shrinkage glazes, kiln cracks, shock crackles and other defects are easy to cause water, salt and pollutants to enter during burial process, causing erosion, or salting out and deglazing after excavation and water drying. Most of these diseases are active and induced diseases, which should be dealt with in time[12].

3.1.3 Avoid excessive treatment of damaged cultural relics:

The main chemical composition of enamels is a silicate, containing Ca, K, Na, Al and other metal elements, and body mainly contains silica and aluminum oxide. The main surface attachment types of marine effluent porcelain are insoluble materials containing Si, Ca, Fe, S and other elements. For the chemical cleaning of the attachment, dilute acid, weak acid and metal complex are often used. These cleansers can also produce certain corrosion to the body and glaze. Mr. Hu Dongbo showed that dilute nitric acid, oxalic acid, EDTA disodium salt, sodium hexametaphosphate and other chelating agent solutions had obvious damage to the glaze when they were cleaned[13].

In order to protect the porcelain body, the cleaning method should be adopted first physical and then chemical, and less acid, alkali and complex should be used. In view of relatively loose covering, generally with scalpel "crescent part" in the way of making a circle to gently remove, and then wipe with a cotton swab to remove the pollutants picked down, observing as you clean, to avoid cleaning excessive damage; For the sediments closely bound to the porcelain body, in the process of removing copper rust with a scalpel, a cotton swab can be dipped into ethanol solution or acetone ethanol solution to soften and erase the sediments in a rolling way. Before chemical cleaning, local experiments should be conducted to screen the materials. The purpose is to give priority to the reaction of the reagent with the target sediments from the perspective of reaction kinetics, and to control the concentration and reaction time, so as to minimize the corrosion of porcelain body and glaze. It is found that the application of EDTA-3Na in the application of softening hard sediments has a better effect, and good concentration and reaction time control will not damage the glaze. The contaminants in the glaze gaps chips and crackles can be removed by applying low concentration oxalic acid solution

or potassium permanganate solution.

3.1.4 Determination of the end point of soluble salt ion removal

The removal of soluble salt, we generally use the combination of deionized water ultrasonic vibration and circulating water spray square method. After a period of desalination, we use a conductivity meter to monitor the change in conductivity, or TDS, in the water. The conductivity of water has a certain relationship with the amount of inorganic acids, bases and salts contained in it. When their concentration is low, the conductivity increases with the increase of the concentration. Therefore, this index is often used to predict the total concentration or salt content of ions in water. TDS (Total dissolved solids) used to measure all the total ion content in water, usually expressed in PPM. The electrical conductivity detector was used to detect the electrical conductivity of the extracted desalting solution, and the test values before and after were recorded and compared, so as to determine whether the desalting end point was reached. When the conductivity value measured for three consecutive times is stable and close to the conductivity of deionized water, it indicates that the desalination end point is reached[14].

Here, of course, the so-called desalination end is a relative concept, this is due to the silicate properties of ceramics themselves and the duration of seawater immersion for hundreds or even thousands of years, so our desalting goal is to reduce the body glaze of soluble salt to a safe value, and desalination soak for a long time will also take away a lot of valuable information, going beyond the limit is as bad as falling short.

3.2 Guidance on the selection of adhesive materials

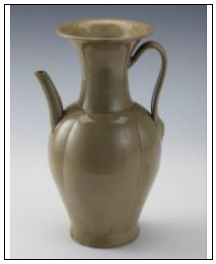
The body of Yue kiln is white and exquisite, a glaze is smooth and clean. According to the principle of minimum intervention and compatibility, the adhesive material must match the strength and performance of the cultural relic material. The specific requirements are high transparency, anti-aging, no reaction with the body and glaze, and the strength and hardness are slightly lower than the porcelain body. A colorless, two-component epoxy resin is commonly used for ceramic bonding. Epoxy resin cured insoluble in most of the solvent, is difficult to take apart according to the rule of reason to place, consider to principles of modern repair concept, can besmear in the first section a solvent-based acrylic resin isolation layer, such as 10% B72 / ethyl acetate isolation layer, dry reoccupy after epoxy resin adhesive, prevent epoxy resin into the section gap is difficult to remove, but for future by ethyl acetate dissolved or low temperature heating to realize apart, to adhesive.

3.3 Guidance on supplementary materials

According to the characteristics of celadon in Yue kiln, the supplementary materials should have low deformation during drying or curing, close to the thermal shrinkage performance of porcelain, certain strength and slightly lower than the hardness of the glaze, not corroding the cultural relic itself and having certain durability. Epoxy resin can be used as filler or finished two-component epoxy resin as filler. The hardness of porcelain fetus is not the same, roughly around 7, glass glaze hardness is more between 5-6.When epoxy resin is used for filling, minerals with a hardness slightly lower than that of porcelain matrix can be used for filling, such as calcite (Moinite hardness 3) and talcum powder (Moinite hardness 1).A small amount of titanium dioxide can be added to adjust the transparency and whiteness, and a small amount of porcelain powder and ore can be added to adjust the hardness and color, so as to achieve the hardness slightly lower than the matrix and slightly lighter than the color of porcelain, so as to meet the material compatibility. The hardness of talc powder is too low, it can be used as a part of the filler in the ceramic epoxy resin complement, not as a single filler in the matrix complement.

3.4 Guidance on the selection of panchromatic and imitation glaze materials

The glaze color of Yue kiln is varied, covered beige, darker bluish gray, grey-brown, sky blue, etc., represent the different colors of divalent iron ion in a different atmosphere. And with the glaze thickness changes, the craft of glazing will also produce changes in glaze color, there will be a slight accumulation of glaze trace at the bottom of the mouth edge. In the panchromatic imitation glaze stage, acrylic coating, water-borne fluorocarbon emulsion or nitro coating that is resistant to aging and not prone to yellowing can be considered [15]. After dilution, multi-layer spraying can be carried out to achieve the light and flowing glaze color. Inorganic pigments with fine particles and bright colors, such as chrome yellow, cyan and iron blue, should be used as far as possible[16]. The natural mineral pigments in the blue-green system are mostly copper containing materials with coarse particles and easy to produce opacification. They are suitable for repairing some celadon varieties with strong opacification in The Yueh kiln. Therefore, it is not necessary to grind mineral pigments to be particularly uniform and delicate. The final stage of the imitated glaze can be finished with a solvent diluted transparent acrylic resin paint, nitrocellulose paint or a more compatible professional imitated glaze coating[17] to give the restored porcelain a texture "like ice and jade". (Fig. 9)



4 Discussion and Prospect

The conservation and restoration of cultural relics is a systematic and step-by-step process of research and operation, which requires heritage conservation workers to constantly explore and learn in their daily work and study. In this paper, we have sorted out the firing defects caused by the traditional porcelain making process of Yue kiln, and on this basis, put forward some understandings and trade-offs of the existing technical means for the conservation and restoration of porcelain cultural relics, so as to point out the direction for future research.

First of all, the concept of conservation and restoration of cultural relics should be clarified, and the basic scale of conservation and restoration should be adhered to, so as not to be interfered by external factors as far as possible, leading to "excessive" restoration. In practical work, we insist that under the premise of ensuring the safety of cultural relics, active and induced diseases should be removed, stable diseases should be avoided as far as possible, some valuable traces of use or burial should be retained, and all traces of production should be retained.

Secondly, before proceeding with the treatment, it is necessary to explore the firing technology of porcelain cultural relics and clarify the difference between the producing area of the kiln and the buried environment, so as to make the conservation and restoration work targeted and effective. For example, the raw materials used for porcelain cultural relics, shaping techniques, loading and firing techniques, etc.; Whether the cultural relics are unearthed or out of the water, if unearthed, whether they are from a cellar or a tomb, the ph value, water content, air permeability of the soil, vegetation roots, microorganisms and other factors must be taken into account. If water is discharged, especially from the sea, factors such as Marine life, seafloor environment (anaerobic or oxygen-rich, whether there is light, fast or slow flow rate, etc.) and salt content of sea water should be comprehensively analyzed.

Third, on the basis of the proper application of existing materials, take the initiative to introduce and develop new cultural and security materials.

It is necessary to constantly select the best among many materials with minimal intervention, and solve the problem by using secure materials and techniques. Of course, the new materials and methods screened out need to pass a series of tests before being applied to the conservation of cultural relics, and they should meet the requirements of reversibility, good aging resistance, low toxicity, easy access and no harm to the cultural relics themselves.

Fourth, we should strive to explore the application of new detection and analysis methods in the cultural and museological industry, summarize and accumulate the basic data applicable to the scientific detection of porcelain cultural relics, and better guide the conservation and restoration work. For example, in practice, we found that inductively coupled plasma mass spectrometry (ICP-MS) and Raman spectroscopy (Ram) can obtain more accurate and meaningful experimental detection results in the analysis of cultural relic composition, which can be used as a supplement and substitute for current common X-ray fluorescence (XRF) and X-ray diffraction (XRD).

Fifth, the preventive conservation of cultural relics must be carried out at the forefront of all cultural relic conservation work, so the study of the best environment for the preservation of cultural relics appears to be of vital importance. Although the environmental requirements of porcelain relics are generally not very strict, the sea water porcelain needs to be considered comprehensively, attention should be paid to temperature, humidity, light, air pollutants and other environmental indicators, to create an appropriate preservation environment. Attaching importance to tracking, monitoring and recording the corrosion rate and change data of cultural relics in various environments can provide scientific support for predicting the life of cultural relics and controlling the development of corrosion.

Cultural relic conservation is a highly interdisciplinary work, which requires the accumulation of knowledge of various disciplines. In daily work, it often faces a variety of thorny problems. As practitioners should consciously study the conditions required for the formation of various diseases. To achieve a comprehensive grasp of ways to mitigate or control the spread of disease and to preserve the health of cultural heritage.

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