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Scientific Research on a Gold- and Silver-Inlaid Bronze Zun from the Han Dynasty

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Abstract: The bronze Zun was one of the more prevalent high-class wine containers of the Han dynasty, representing the highest level of decoration in bronze at the time. However, little has been reported about its technical characteristics and scientific value. In this paper, the samples were selected for analysis based on scientific analysis, following the principle of "minimal intervention", and a bronze Zun from the Han dynasty in the Gansu Provincial Museum collection was studied using ultra-deep field microscopy, X-ray flaw detection, X-ray fluorescence spectrometry (XRF), scanning electron microscopy (SEM), and energy spectrometry (EDS). The results show that the gold and silver decoration on the bronze is inlaid rather than gilt. Secondly, the body and lid of the vessel are molded in one shot, with the bird-head-shaped and animal-foot-shaped components cast separately and then attached to the lid and body. Thirdly, the corrosion of the bronze Zun is characterized by the copper matrix being corroded first and most severely, followed by the silver and, finally, the gold. The high purity of the gold wire embedded in this bronze Zun, the fine width of gold wire (154–190 µm), and the magnificent decoration show the excellent processing technology level of the precious metal and the high aesthetic level of ancient man during the Han dynasty. The results of the analysis of this bronze Zun can provide an essential reference for research on bronze vessels of the same type, the techniques of gold and silver misalignment, and the development of the history of bronze manufacture and technology during the Han dynasty.

Keywords: Han dynasty; bronze Zun; gold and silver inlaying; scientific research

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1. Introduction

The Zun (bronze vessel) was a practical everyday wine vessel that appeared during the Warring States period (476–221 BC) and flourished during the Han dynasty (202 BC–8 AD). It came in two shapes similar to basins or barrels, mostly with three feet or a ring foot [1–3]. The bronze Zun was a more expensive, high-class wine container during the Han dynasty and was one of the most prominent wine containers. In ancient China, there was a tradition of using wine to entertain guests, express personal feelings, and even treat diseases. China's wine culture began almost along with the splendid civilization of 5000 years [4]. Because of this penchant for wine, the imperial aristocracy was very particular about wine vessels under the social conditions of the time. As a result, the popular Zun during the Han dynasty was particularly prestigious at the time, representing the Han bronze wares' highest level of decoration.

The bronze Zun of the Han dynasty enjoyed unprecedented development and reached a high level of decoration based on the techniques used to make it. Standard decorative techniques of bronze ware include inlay [5,6], gilding [7], gold inlaying [8], gold wrapping [9], burin engraving [10], and so on. However, because of the paucity of surviving ancient texts, there is still further research required into gold inlaying [11]. From the excavated cultural relics, we can understand that the ancient misprinted gold and silver process includes two forms of gold inlaying and gilding [12]. Inlay uses gold wire, gold plates, silver wire, silver plates, and other decorative objects on cast bronze inlaid with various ornaments or inscriptions. The gilding system, sometimes called the "gold coating method", uses gold amalgam on the surface of the bronze, which is repeatedly coated with gold; after the mercury evaporates, the gold remains on the object's surface [13]. In connection with this, gilded objects decorated with gold may also have gold inlaying, and those gilded with silver may also have silver inlaying. The process of gold and silver inlaying is more complicated than gilding and requires large amounts of gold and silver, so gold- and silver-inlaid bronze is precious; studying such objects is of significant value to improve our understanding of their craftmanship.

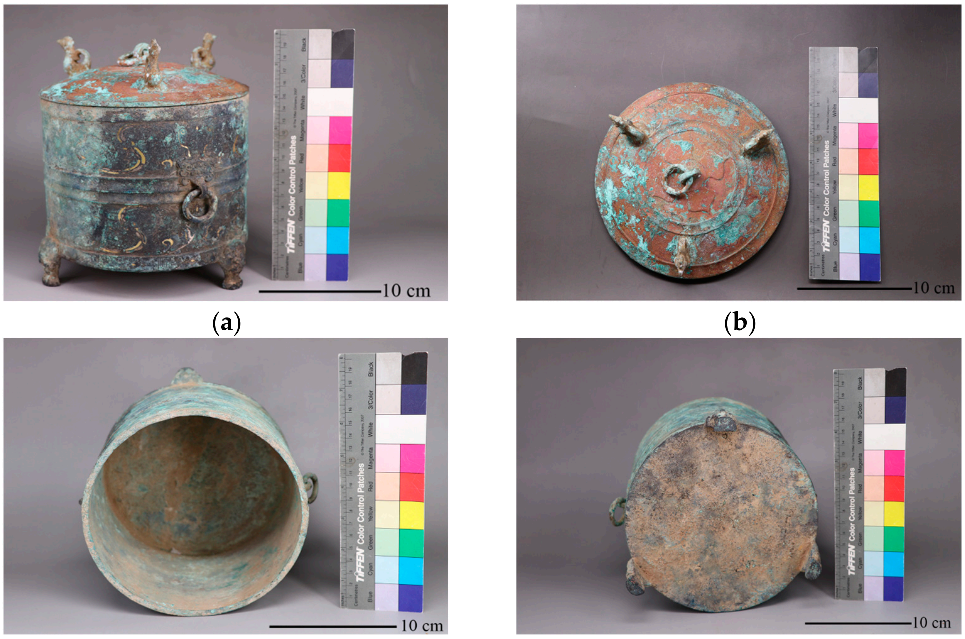
There are some studies on the gold and silver inlaying and gilding processes on bronze at home and abroad. Based on the available research, the basic steps of the gilding method used in different regions are approximately the same [14–17]. A study by Oddy et al. on the composition of decoration on gold vessels, silver vessels, and bronze vessels from the ancient and medieval periods found that the statue of the Roman emperor Niello (1st century AD) was made of bronze. At the same time, the object's surface was inlaid with silver and had a shallow groove with a black coating, known as "Niello". The technical analysis carried out by Konstantinidi-Syvridi E. and others, based on archaeological and experimental data, revealed that a decorative technique using gold inlay on the surface of luxury weapons, called gold embroidery, was prevalent in the early Mycenaean period [18]. This metalworking technique involved the dense placement of tiny gold wires or particles of gold (also known as gold bars) next to each other and, finally, engraving them into a decorative pattern. Although this is a gold inlaying technique, it differs from what we have described as gold inlay. It is worth noting that the dimensions of the bars used in this decoration technique range from 0.3 to 8 mm in length, 0.4 to 1.5 mm in width, and 0.3 to 0.6 mm in thickness, respectively, a clear difference from Chinese Han dynasty metalworking techniques. Yang et al. studied the gilding process of a Warring States bronze chime from the Sichuan Museum collection. They speculated that the gold wire of chime bells was decorated with groove-gilding technology [19]. Liu et al. carried out the conservation and restoration of a gold-inlaid silver belt hook excavated during the Warring States period based on modern instrumental analysis methods [20]. However, the available research findings have yet to explore the depth of the gold and silver inlaying process on bronze Zun.

The conservation of cultural relics is not only about treating the damage but also about extracting as much information as possible about the relics and digging deeper into their artistic, historical, and scientific values [21]. Based on the above, we have undertaken a scientific analysis of a Han dynasty gold- and silver-inlaid bronze Zun from the Gansu Provincial Museum collection to provide a reference for research on the gold inlaying process of similar types of bronzes and the scientific and technological history of bronze production during the Han dynasty.

2. Materials and Methods

2.1. Materials

Figure 1 shows the appearance of the gold- and silver-inlaid bronze Zun. It is cylindrical, with a lid, a flat base, and three animal-shaped feet. The overall weight of the object is 2.1 kg, of which the lid weighs 0.54 kg and the body weighs 1.56 kg. The overall height is 20 cm, of which the body is 15 cm high and the lid is 5 cm high. The outer diameter of the body is 16.6 cm, and the inner diameter of the mouth rim is 14.2 cm. The top of the lid is surmounted by a looped handle, with three buttons outside the loop, and the central area is carved in relief with a persimmon peduncle, with symmetrical classes with a ring handle on the belly. Neither the lid nor the base of the vessel is decorated or inscribed. In addition, the object's surface is covered with a green and black patina due to its burial and preservation environment, which has caused some of the decorative motifs on the vessel's surface to be covered. It is tentatively determined that the wires removed from the body of the bronze Zun and lid are gold and silver.



(**a**) (**b**) (**c**) (**d**)

Figure 1. Appearance of the bronze Zun. (**a**) is overall appearance; (**b**) is lid appearance. (**c**) is internal body. (**d**) is bottom of the body.

2.2. Methods

The analysis and sampling for this study were carried out using non-destructive or minimally invasive methods. Therefore, a digital VHX–6000 microscope (KEYENCE, Osaka, Japan) was used for microscopic observation. The bronze vessel was photographed fluoroscopically by Smart Evo 300D X-ray flaw detection (YXLON, Hamburg, Germany) to observe its internal structure and the information on the surface decoration covered by rust without special treatment of the samples. Non-destructive testing of the alloy composition of the bronze Zun was performed using X-ray fluorescence spectroscopy (Thermofisher, Waltham, MA, USA). The body was examined in the normal metal mode, and the gold and silver wires were examined in the precious metal mode with a test time of 60 s. The microscopic morphology of the samples was observed by scanning electron microscopy (SEM) (JSM–6610LV, JEOL, Tokyo, Japan), and the matrix and inclusions of the samples were analyzed by X-ray spectrometry (INCA X-ACT 250, Oxford Instruments, High Wycombe, UK) for their micro-zone chemical composition.

3. Results and Discussion

3.1. Ultra-Deep Field Microscope Analysis

Figure 2a shows the gold wire coming off the edge of the lid of the Zun and the exposed grooves, which would have been formed by a chisel. It can be seen that the burin grooves are wide on the outside and narrow on the inside, which is different from the usual grooves in decorated gold, which are wide on the inside and narrow on the outside. The different widths and depths of the chisel grooves also illustrate the random nature of the ancient artisan's chiseling operation. The gold filigree is essentially flaking off as a whole with little residual trace, and it can be tentatively inferred that this would not have been formed by the gilding method of gold amalgam. In addition, measurements show that the width of the gold wire varies between 154 and 190 μm (Figure 2b), which is much smaller than the size of the gold used in the aforementioned Mycenaean weapons inlaid with gold, indicating the high level of metalworking technology of the Han dynasty [18]. Figure 2c shows a detailed view of the articulation of the gold wire, which can be seen at the junction where the wire is overlapped, with apparent traces of jointing work, which may be a remedial measure for the artisan's underestimation of the length of the wire when it was inlaid and which also indicates the use of smashing and pounding in the process of inlaying the gold wire. The uneven edges of the gold can also be seen, with traces of cuttings. As can be observed in Figure 2d, there are clear, congruent polishing scratches on the gold filigree's surface, which the whetstone grinding created. In addition, the edge of the chiseled groove at the location of the gold and silver is tilted inward, presumed to be caused by hammering to make the malleable gold and silver pieces more closely connected to the chiseled groove.

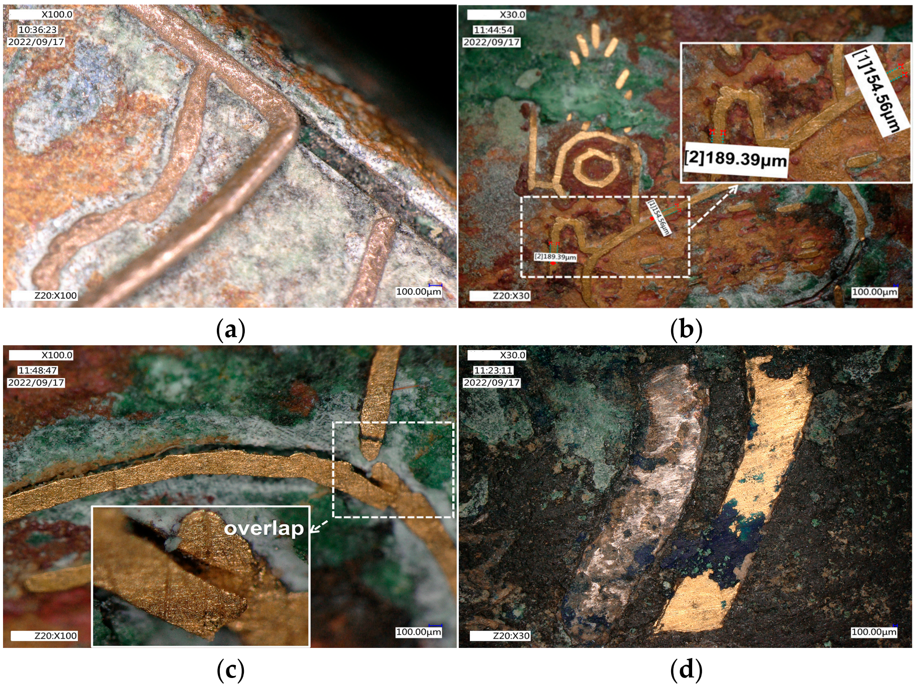


Figure 2. Ultra-deep field microscope photos of the bronze Zun: (**a**) the chisel groove; (**b**) the gold wires ([1] and [2] refer to the width of the gold wire measured at 2 different locations.); (**c**) the junction of gold wires; and (**d**) the surface grinding marks of the gold and silver pieces.

3.2. X-ray Flaw Detection Analysis

In order to gain further insight into the overall manufacturing craft of the bronze Zun, it was tested and analyzed using X-ray flaw detection imaging inspection techniques, the results of which are shown in Figure 3. The delicate ornamental motifs and decorations on the lid and body can be seen on the X-ray flaw detection graphs. The surface of the lid is decorated with bird and animal motifs as the prominent motifs, and it is decorated with a combination of cut diamond-shaped pattern pieces and gold thread to form auxiliary motifs. The main decoration and auxiliary decoration patterns on the surface of the bronze Zun are clearly layered, which is a typical pattern of decoration in the Han dynasties [22]. The bronze Zun is decorated with a bow-string pattern, a diamond-shaped pattern, and wavy patterns around it, with three animal-head-shaped components at the base to provide support (Figure 3a,b). Due to the photographic limitations of X-ray flaw detection, it is difficult to distinguish the patterns of the front and back sides of the bronze Zun with cavity structure, and the decorative motifs around the body are superimposed onto the same plane while taking an X-ray film. Despite this limitation, fine surface decoration and motifs can be observed (Figure 3b). The "SS" decoration on the body is also visible in Figure 3b, consisting of cut gold and silver filigree. The shades of color of the "SS" motifs in the X-rays vary, and it is speculated that the darker and lighter motifs are silver and gold, as the higher the atomic mass and the higher the density, the lower the blackness of its position [23]. This is consistent with the results of the exterior photographs of the objects (Figure 1a) and the super-field microscopic analysis (Figure 2d), all of which demonstrate the fine line engraving and delicate decorative techniques that were part of the superb handwork of ancient bronze decoration. However, varying degrees of detachment and loss of gold and silver filigree and flakes can also be observed.

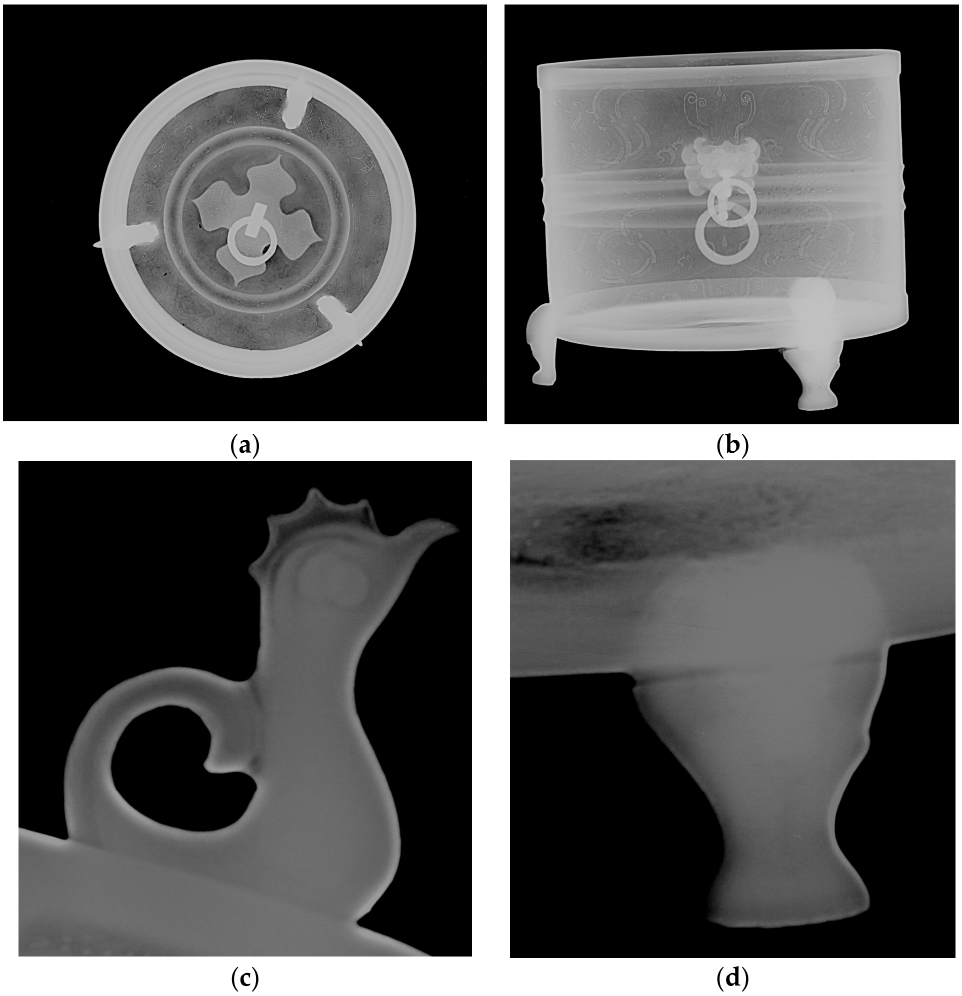


Figure 3. X-ray flaw detection graphs of the bronze Zun: (**a**) the lid of the bronze Zun; (**b**) the body of the bronze Zun; (**c**) the bird-head-shaped component on the lid; and (**d**) the animal-foot-shaped component on the body of the bronze Zun.

From Figure 3a, the lid has a relatively dense texture, indicating a relatively homogeneous copper liquid during the casting process and a stabilized subsequent cooling process. Four shading spots on the lid are presumed to be possible shrinkage holes created during the casting process [24]. This is because, during the solidification and shrinkage of the coppery liquid, the thinner-walled locations would have solidified first, thus blocking the passage for the flow of the coppery liquid. The subsequently solidified locations would not have been able to replenish the coppery liquid as they continued to cool and shrink, resulting in air pockets and shrinkage holes.

In addition, the difference in the degree of X-ray absorption between the bird-head-shaped components at the top of the lid and the body of the lid and between the animal-foot-shaped components and the main body of the vessel should be due to the different densities of the materials (Figure 3c,d) [25]. Combined with the appearance photos of the lid and the body (Figure 1), no sign of a match was observed, and it is presumed that molding was used in divided model-making. Therefore, it seems as though the main body of the bronze Zun was cast once, and the three bird-head-shaped and three animal feet of the lid were cast separately and then connected.

3.3. X-ray Fluorescence Spectroscopy Analysis

The lid base, the body, the bird-head-shaped components, and the animal-foot-shaped components of the bronze Zun were selected for surface alloy composition testing. Table 1 presents the percentage content of the elements and the respective standard deviations (RSDs) obtained in both bronze samples. The main elements in this bronze Zun are copper, tin, and lead, with 73.28%, 21.03%, and 3.05% in the base of the lid and 77.00%, 17.66%, and 1.91% in the base of the body, indicating that the bronze is a ternary Cu-Sn-Pb alloy and a high-tin bronze [26]. The copper, tin, and lead contents of the bird-head-shaped lid and the foot of the animal on the body are 66.55%, 18.12%, 1.91%, 80.63%, 15.89%, and 0.64%, respectively. The difference between the elemental content of the base of the lid and that of the body of the bronze Zun confirms, on the one hand, the findings of the X-ray analysis that the bronze Zun and the lid would have been cast in one piece and that the bird-head-shaped and animal-foot-shaped components were cast separately and then attached to the lid and body. On the other hand, the rust on the surface of this bronze may have interfered with the XRF results to some extent.

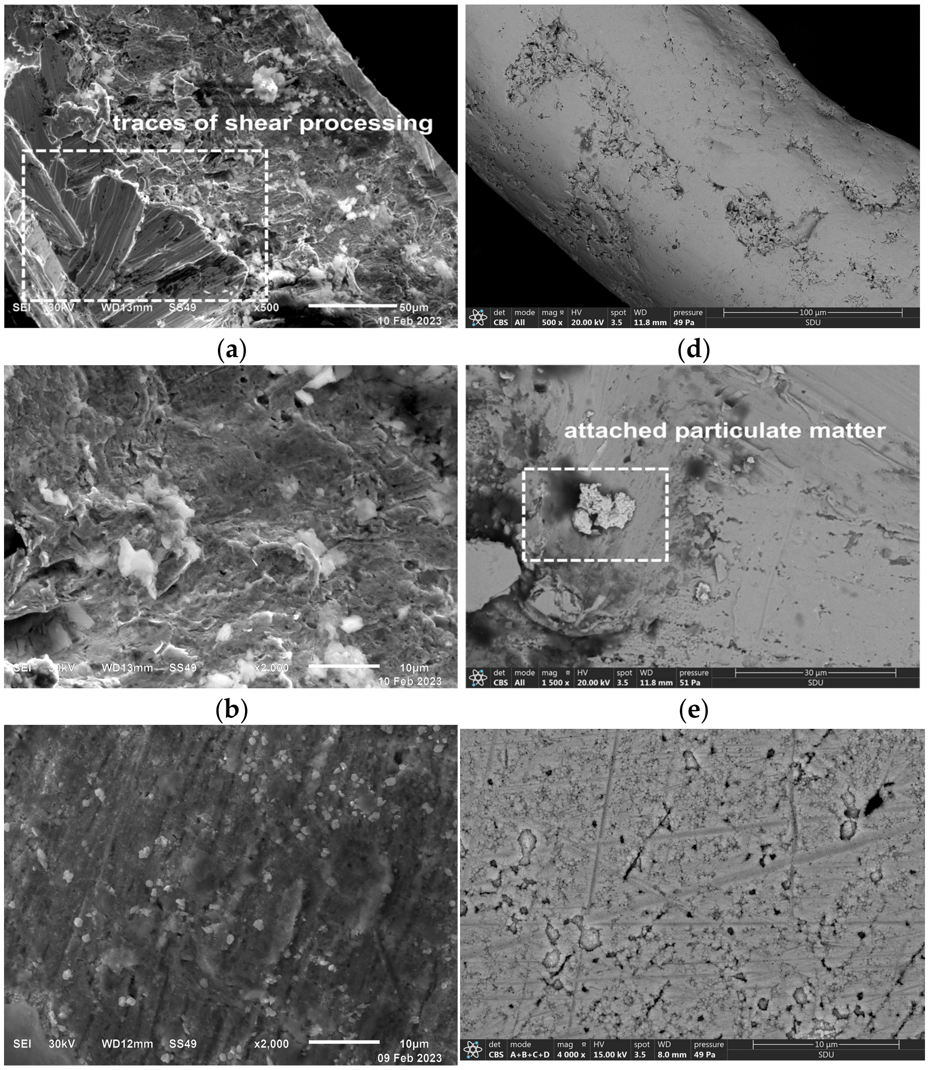
Table 1. XRF results of each part of the bronze Zun (wt %).

No.	Test Content	Fe	Cu	Pb	Sn
1	Lid body	0.07	73.28	3.05	21.03
	(RSD%)	(21.02)	(2.73)	(11.52)	(9.57)
2	Bird-head-shaped components	0.51	66.55	1.92	18.12
	(RSD%)	(17.93)	(3.97)	(13.66)	(11.73)
3	Vessel body	2.90	77.00	1.91	17.66
	(RSD%)	(11.25)	(3.82)	(13.98)	(12.11)
4	Animal-foot-shaped components	2.41	80.63	0.64	15.89
	(RSD%)	(10.49)	(2.35)	(18.01)	(12.56)

3.4. SEM–EDS Analysis

Figure 4 shows the SEM images of the gold and silver wires. SEM micro-zone analysis reveals traces of shear processing on the surface of the gold wire, a process that was used to better embed it into the surface of the bronze (Figure 4a,b), with impurities adhering to the surface. After initial cleaning of the cross-section of the gold wire, it can be seen that the entire profile is relatively homogeneous, with tiny particles of gold (Figure 4c). Meanwhile, an uneven surface of the silver wire can be observed (Figure 4d), and particle attachment is also evident from the center of Figure 4e. The profile of the silver wire shows a more uniform distribution, similar to that of the gold wire (Figure 4f). This is because both gold and silver have the same face-centered cubic crystal structure, as well as similar cell parameters and chemical properties [27].

The analysis results of the surface and profile elements of the gold and silver wires and their relative standard deviations are shown in Table 2. The surface of the gold wire contains 67.48%, 7.68%, 3.61%, 4.42%, 1.40%, 14.86%, and 0.91% of gold, silver, copper, tin, chlorine, oxygen, and aluminum, respectively. The gold wire profile contains 92.50% gold and 7.50% silver, which can be inferred to be pure gold. This is because gold in its natural state, whether obtained from a placer or vein gold mine, usually contains considerable amounts of silver (usually 5.00% to 45.00% by weight) and other major associated impurities of gold [28]. Some scholars believe that the small amount of silver detected in the gold is more likely to represent a natural impurity in the gold than an artificially prepared alloy [29]. However, to conclude that the gold used for decoration in this bronze is natural gold smelted or synthesized through the later artificial addition of silver, more bronze or gold and silver vessels of the same type need to be analyzed.



(**a**) (**d**)
(**b**) (**e**)
(**c**) (**f**)

Figure 4. SEM photos of the gold and silver wires: (**a**,**b**) the surface of the gold wire; (**c**) the gold wire profile; (**d**,**e**) the surface of the silver wire; and (**f**) the silver wire profile.

Table 2. SEM–EDS results of the composition analysis of the gold and silver wires.

Test Content	wt%								
	Au	Ag	Cu	Sn	Cl	S	Si	Al	O
the surface of the gold wire (RSD%)	67.48 (9.52)	7.68 (10.76)	3.61 (11.43)	4.42 (11.21)	1.04 (13.20)	-	-	0.91 (15.76)	14.86 (9.67)
the gold wire profile (RSD%)	92.50 (5.33)	7.50 (9.47)	-	-	-	-	-	-	-
particulate matter on the surface of the silver wire (RSD%)	-	99.60 (3.22)	0.40 (16.87)	-	-	-	-	-	-
particulate matter on the silver wire (RSD%)	-	11.00 (11.73)	71.00 (7.59)	16.00 (10.56)	1.40 (13.61)	0.30 (17.24)	0.20 (18.57)	0.10 (19.0)	-
the silver wire profile (RSD%)	-	78.30 (8.12)	-	-	21.7 (10.71)	-	-	-	-

Furthermore, the Cu and Sn elements contained on the surface of the gold wire would have come from the body of the bronze Zun. The traces of Cl and Ag elements detected on the surface of the gold wire could have been caused by the corrosion of the silver contained in the gold wire, which is probably in the form of silver–chlorine species [30,31]. On the one hand, the silver content on the surface of the silver wire is 99.60% silver and 0.40% copper. The silver, copper, chloride, and tin contents of the particulate matter on the surface of the silver wire are 11.00%, 71.00%, 1.40%, and 16.00%, respectively. The detected copper element would result from the silver wire being embedded in the bronze and prolonged contact with the bronze matrix of the vessel body. On the other hand, the main constituent elements of the energy spectrum are O, Ag, Cl, and Cu, and it is presumed that the particulate matter is mainly cuprous oxide and silver chloride. The silver wire profile of 78.30% silver and 21.70% chloride suggests that bronze is more severely corroded.

By comparing and analyzing the elemental composition and content of the profiles of gold and silver wire, it was found that the content of gold was higher than that of silver, and the silver wire was corroded a little more seriously. Combined with Figures 1 and 2, the copper matrix's corrosion was more severe than that of the silver wire, with more and thicker corrosion at the gold and silver inlay decoration. Because the gold- and silver-inlaid bronze Zun mainly contains Au, Ag, and Cu, three different metals, these three metals' electrode potentials are different ($E_{Au} > E_{Ag} > E_{Cu}$). According to electrochemical principles, it is known that when there is an electrolyte in a buried or preserved environment, the copper matrix is corroded first, followed by silver, and finally gold. Suppose the patina is treated directly with chemical cleaning agents on the one hand. In that case, this will result in the surface of the bronze being slightly lower than where the gold and silver are decorated, affecting the artifact's appearance. On the other hand, the cleaning solution may even enter between the gold and silver filigree and the bronze matrix, dissolving the patina therein and possibly causing the decoration to loosen or fall off. If the mechanical method removes the rust, it may leave scratches on the surface of the gold and silver decoration due to its soft texture [32]. During the conservation and restoration of the relics at a later stage, the pros and cons of these two methods should be weighed, and a reasonable conservation program should be developed.

In addition, Ag is susceptible to corrosion into AgCl, and this corrosion product can migrate to all parts of the object and even contaminate the surface of the artifact [33]. The absence of mercury in the SEM–EDS results leads us to conclude that the gold wire in this bronze Zun was not formed by the fire-gilding method, which supports our discussion in Section 3.1.

4. Conclusions

This study is based on morphological and chemical elemental analyses of a bronze Zun from the Han dynasty in the Gansu Provincial Museum. Through comparative analysis of the test results, the main conclusions were as follows:

(1) The gold and silver inlay decorations on this bronze Zun used techniques such as chiseling grooves, inlaying, and polishing rather than gilding;
(2) The body and lid would be cast in one piece, with the bird-head-shaped and animal-foot-shaped components cast separately and then attached to the lid and body;
(3) The bronze Zun, being a gold- and silver-inlaid bronze vessel, is characterized by the corrosion of the copper matrix first and most severely, followed by the silver, and then the gold;
(4) The high purity of the gold wire embedded in this bronze Zun, the fine width of gold wire (154–190 μm), the large amount of gold and silver used throughout, and the magnificent decoration reflect the abundance of gold minerals during the Han dynasty and the widespread dissemination and development of gold processing techniques during this period. At the same time, the technical and aesthetic level of precious metal processing is high. However, to determine whether the gold used in the decoration of this bronze is natural gold smelted or synthesized by the later

addition of silver, we believe that more bronze or gold and silver vessels of the same type need to be analyzed.

The analysis of this gold- and silver-inlaid bronze Zun is of great importance to studying human technology and lifestyle during the Han dynasty, as well as providing an essential reference for the conservation and restoration of similar bronze objects and the study of technology.

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