Metallurgical Characterization of a Copper-Alloy Aramaic-Inscribed Object from Tulûl Masʿud (Elyakhin)

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Abstract: An Aramaic-inscribed object made of copper-alloy was discovered in 1993 in the southwestern part of Tulûl Masʿud (Moshav Elyakhin) and has recently been studied using an archaeometallurgical approach. Based on visual testing and multifocal light microscopy observation, the object was probably produced in a nearby workshop, with the inscription engraved using a sharp tool during the production process. Given the larger assemblage of inscribed copper-alloy artefacts from the site, this item appears not only to have been used as a cultic object, but was also most probably made for the purpose of cultic offerings. The XRF analysis results of the Aramaic-inscribed object after it was sanded revealed the core metal to have been made of relatively pure copper with a tin content of less than 1.0 wt. % Sn. The choice to produce the object using a low-tin copper-alloy indicates that the alloy was chosen based on technological considerations, in order to facilitate plasticity in fashioning the part into its cylindrical shape. The manufacturing process involved bending the object while it was hot and shaping it into its final form by means of several cycles of forging and annealing. Although the current research has revealed the bulk composition and the general manufacturing process of the object, the microstructure of the core alloy could not be observed because destructive testing was not permitted. Although only a single copper object was analysed, the current archaeometallurgical study allows to gain further information on metallurgical knowledge and manufacturing processes of copper objects in the Persian period Levant.

Keywords: Aramaic-inscribed; archeometallurgy; copper-alloy; manufacturing process; non-destructive testing (NDT); Persian period; Tulûl Masʿud (Elyakhin)

1. Introduction
1.1. General Description of the Discovery of the Incised Aramaic Inscription

A copper-alloy object with an incised Aramaic inscription was discovered by chance in 1993. An introductory article presenting the main characteristics of this find, focusing on its historical context, epigraphic characteristics, and other finds from the site, is to be published in `Atiqot [1] (Lewis et al. in press). The present article focuses more on the object’s metallurgical characteristics. The object was found on the surface, about halfway up the eastern slopes of the hill of Tulûl Masʿud, located in the south-western part of Moshav Elyakhin in the central Sharon plain, Israel. Shortly after its discovery, the object was given to the Israel Antiquities Authority (IAA). In 2021, it was relocated to the IAA conservation lab for study and publication.
1.2. The Site of Tulûl/Tell Mas’ud

The site of Tulûl Mas’ud or Tell Mas’ud [2] (Sheet XI) (701596/192556 (New Israel Grid)) is located in Emeq Ḥefer, in the central Sharon plain, on a red loam hill (44 AMSL) that dominates the landscape in the south-western part of Moshav Elyakhin (Figure 1).

The site was badly damaged in the mid-1950s, after being levelled by a bulldozer as part of construction operations to install a concrete water tank on top of the hill (Figure 2). The hill of Tulûl/Tell Mas’ud was frequently mentioned by archaeological surveyors in the late 20th century as a possible location of a Persian-period temple, but was never excavated.

The surveyors’ work yielded finds such as an ashlar stone wall; pottery dated to the Late Iron Age, Persian, Hellenistic, Roman and Byzantine periods; a limestone statue with a plank-shaped body; an object made of alabaster, a mortarium; an Irano-Scythian arrowhead; and a black stone statue of Osiris with an hieroglyphic inscription on its back, dated to the Hellenistic period ([3] (Tulûl Mas’ud 14-20/21); [4] (p. 10); [5] (pp. 138–140); [6] (p. 13); [7] (p. 144); [8]; [9] (site no. 94, see also sites nos. 84–86); [10] (pp. 118–123); [11] (pp. 153–158)).
Unfortunately, archaeological surveyors were not the only visitors to the area, as several artefacts from this site have also found their way into private collections: five bronze bowls, a bronze cymbal, and a bronze fragment, possibly from an oil lamp, all of which bear Phoenician and Aramaic script inscriptions [12] (pp. 69–89); 15 Sidonian, Tyrian, and Samarian silver coins from the late 5th and especially 4th century BCE [13] (pp. 17–20); and other finds such as fibulae and arrowheads, as well as gold-, silver-, and bronze-made jewellery [12] (p. 69).

The site is still undergoing significant development and traces of modern construction activities can be seen in many areas.

Figure 2. An oblique aerial photograph of Tulūl Mas‘ud (Elyakhin), looking north-west (photo by R.Y. Lewis).

1.3. The Object: Technical Description

The tube-shaped object that is the focus of the present study (Figure 3) is 8.5 cm long and 3.6 cm in diameter. It weighs 176.8 g (Precisa BJ 2200C electronic scales, max = 2200 g and d = 0.01 g).

A small hole (3 mm in diameter with a 2 mm wide depression) appears close to the end of the rounded front of the tube, probably for a nail to secure it to a handle (Figures 3a and 4c). The thickness of the plate comprising the tube is 2 mm at the shaft and 4 mm at the flattened broken end. The plate that makes up what appears to be the front of the tube is rounded, while the back is hammered with one side of the plate partially folded over the other side where they meet. A two-line inscription was incised (of which only part has survived). Both inscribed lines are nearly 8 cm in length, with the upper line of letters reaching a maximum width of about 1 cm and the lower line reaching a maximum width of 1.5 cm. The copper-alloy tube (Figures 3 and 4) seems to have been used as part of a larger object (perhaps a fire pan or an incense shovel) or to have been the handle (or part of it) of a ceremonial weapon whose incised inscription dedicates it to the divinity/divinities of a sanctuary. As the head of the larger object is missing, we cannot determine unequivocally the type of object to which the copper-alloy tube was originally attached.
The post-Iron Age corpus of Aramaic lapidary inscriptions was recently summarized by Lemaire [14]. Although the term “lapidary script” is usually applied to inscriptions on stones used for official purposes, it is also used to describe inscriptions on other materials [15].
Among the Aramaic lapidary inscribed metal objects of the Persian period found at various sites, those published from the Elyakhin site [12] (pp. 69–89) can be taken as reference material, given their provenance and the fact that most of them feature Aramaic script of the Persian period and refer to the same divinity, namely ‘strm.

The inscription on the object reads as follows:

1. [$\ldots$] גברט made [$\ldots$

2. [$\ldots$] To ‘strm’ [$\ldots$

The script of the inscription was discussed in some detail by Lewis et al. (in press [1]), as were its prosopography and syntax. The offering to the divinity ‘strm’, meaning “to the god/dess/es ‘strm’”, can be compared with the seven other Elyakhin Aramaic inscriptions [12] (pp. 73–83). The goddess ‘štrt was the principal Phoenician-Punic female deity in the first millennium BCE [16]. In regard to the -m ending of ‘strm, Zadok [17] (pp. 34–38) suggested that this is a plural form. Lipiński, in his study of the cult of ‘Ashtarum in Achaemenian Palestine, considered it to be ‘Ashtar’s name with a mimation, to be
compared to the Ammonite divine name Milkom [18] (p. 315). Lipiński also argued for a North-Arabian origin, supported by his new readings and interpretation of the Elyakhin Aramaic bronze bowl’s Aramaic inscription no. 6 [12] (pp. 81–83); see also [19]; [20] (pp. 312–313, note 22). Based on the palaeography of the inscriptions and the coins recovered from the site [13], our object may be dated from the end of the 5th to the mid-4th century BCE.

2. Metallurgical Background

Copper was one of the first and most widely used metals during antiquity. According to the literature, native copper had been in use since the 8th millennium BCE [21]. It was heated to a temperature of a few hundred degrees Celsius and then worked and shaped by hammering [22]. The production of molten copper from its ore by means of a smelting process started around 4000 BCE [21,23]. During antiquity, copper was smelted in a furnace from copper-oxidic and sulphidic ores [24]. Copper smelting technologies have also been recognized as having taken place along the Levantine coast during the 6th millennium BCE [21,25]. Ancient copper smelters and smiths knew how to select ores and manufacture products made of high-quality copper alloys, with a composition that would be appropriate for the production of certain artefacts designed for particular applications [26]. Iron, arsenic, and antimony are among the most common impurities in ancient smelted copper, depending on the smelting method used for the minerals and the temperature of the furnace [24].

Copper is easily cast and the addition of 10 weight percentage (wt. %) of tin to copper reduces the melting temperature from 1084 °C to around 950 °C and increases the hardness of the alloy [21,27]. Evidence of early copper casting in the 5th millennium BCE was discovered in Varna, Bulgaria [21]. As-cast products made of copper alloys typically exhibit a dendritic microstructure and generally tend to display cast imperfections such as interdendritic porosity and spherical porosity, which results in gas bubbles being captured in the solidifying cast metal [24,26,28]. Arsenic and tin tend to reduce the formation of cast bubbles and porosity in copper artefacts [21,28].

During antiquity, bronzes made of relatively pure copper and low tin were used for different applications based on their excellent ductility, ability to enhance strength and strain-hardness, and the good corrosion resistance of the metal [29]. Ancient objects made of copper alloys were generally either as-cast or cast and worked products [30]. Copper parts were often shaped by cold hammering and annealing cycles. When a copper alloy is worked by plastic deformation at temperatures much below its melting point, it becomes stronger because of the increment in dislocation density, resulting in strain-hardening [31]. Annealing treatment restores the lost ductility and thus permits further deformation of the alloy [31,32]. The microstructure of archaeological copper artefacts results from various factors, including the objects’ composition (alloying elemental concentration), cooling rate during the casting process, presence of impurities such as inclusions, and the operation of additional thermal and/or mechanical treatment [30].

Chemical analysis combined with microanalysis can help to reveal the production process of copper-based objects and occasionally also to uncover the provenance of the raw material. The technology used to produce copper through plastic deformation and annealing cycles can be best observed by means of metallographic analysis, revealing the microstructure of an object [22]. However, because of the rarity of ancient copper artefacts, it is often not possible to examine metal items using a destructive test such as a metallographic analysis [22,32]. In such cases, the minimally destructive novel technique of field multi-focal metallography (FMM) can be employed [33]. Moreover, understanding the chemical composition, technological skills, and production abilities of the metal objects within a particular region of human settlement will in the long run lead us to a greater understanding of the economic trends as well as of the social and cultural interactions of ancient civilizations [22].
The best way to identify native copper is by performing distinctive metallographic analysis and observing the metallographic specimens under light microscopy (LM) and scanning electron microscope with energy-dispersive spectroscopy (SEM-EDS), using both secondary electron (SE) mode and back-scattered electron (BSE) mode. When such an analysis is performed, native copper can be recognized by its high purity. However, a quality smelting process of copper from copper ores (such as malachite) can result in copper metal as pure as native copper. Therefore, because it can be rather challenging to differentiate between native copper and metal smelted from copper ores by means of elemental analysis, it is also recommended to perform trace element analysis using techniques such as inductively coupled plasma mass spectrometry (ICP-MS), laser ablation ICP-MS (LA-ICP-MS), neutron activation analysis (NAA), atomic absorption spectrometry (AAS), as well as X-ray fluorescence (XRF) analysis [22,34].

Because domestic tools commonly did not require high strength, they were frequently made of low-tin bronzes [26]. Copper objects were constantly recycled owing to their valuable nature [21,26]. Hence, in antiquity, metal scraps were commonly melted and re-cast [21]. Ancient bronzes with up to about 2 wt. % Sn usually indicate the use of recycled metal [31,32]. At a low-tin content of less than 5 wt. % Sn, it is possible (depending on the cooling rate and the type of casting involved) that all of the tin will be absorbed into the dendritic growth. If the cooling rate is sufficiently slow, the amount of interdendritic δ (Cu₃Sn₈) phase will be reduced dramatically or even disappear completely, resulting in α phase only. Nevertheless, at a tin content of around 10 wt. %, it is quite rare to achieve total absorption of the δ phase, hence the dendrites will be surrounded by a matrix composed of both α + δ eutectoid. As the tin concentration rises, the amount of interdendritic eutectoid also rises [28].

Ancient copper artefacts are often discovered in a relatively good state of preservation owing to their excellent resistance to corrosion. However, under some aggressive environmental conditions, copper and its alloys will suffer from general or local degradation following a long-term corrosion process [24]. Examination of ancient objects made of copper alloys found at various archaeological sites in the Mediterranean region have shown that their patina layers had been enriched by different soil elements, such as Si, Fe, Al, P, S, Ca, and Cl, owing to the interaction between the soil elements and corrosion products [31,35].

3. Experimental Methods and Tests

A non-destructive testing (NDT) of a Persian-period copper-alloy tube decorated with Aramaic inscription was performed in order to cause minimal damage to the object. The examination included visual testing (VT), handheld XRF (HH-XRF) measurements, and multifocal LM observation.

(a) VT inspection was carried out to examine the state of preservation of the object and to detect macroscopic level details that can indicate the manufacturing process.

(b) HH-XRF chemical analysis was carried out using an Oxford X-MET8000 instrument with silicon drift detector, equipped with a 45 kV Rh target X-ray tube. Each measurement was performed for 30 s on a detected area of 5 mm in diameter, inside a bench-top stand. The HH-XRF instrument was calibrated with standard calibration samples made of copper and copper-bronze (11.1 wt. % Sn). Such instruments can measure alloy elements to within an accuracy of up to 0.5% of the measured value. The HH-XRF analysis of the object’s exterior surface may not be representative of its bulk composition owing to the thick patina layer and surface corrosion. The elemental composition of the patina layer and corrosion products can be enriched in certain trace elements and is often depleted in copper concentration in comparison with bulk metal. The reliability of the measurements can be improved by removing the patina layer and corrosion products [22,29]. Consequently, the external surface of the object was roughly sanded using a 320 grit silicon carbide abrasive paper to
expose the bulk metal and then cleaned with ethanol. The HH-XRF approach presents a challenge in differentiating between the peaks of As and Pb, owing to the low level of peak overlap. The HH-XRF measurements were thus performed through a comparison of two independent peaks: As Kα was compared to Pb Mα and As Kβ peak was compared to Pb Lβ [29].

The surface of the object was observed using a HIROX RH-2000 digital multifocal LM equipped with powerful software, encoded optics, high-intensity LED lighting, and a light sensitivity sensor at high-resolution HD (1920 × 1200), presenting high pixel density and low image noise. This advanced tool allows the following of surface colours, topographies, and morphologies [36].

4. Results and Discussion

An 85 mm long Persian-period copper-alloy tube, probably the handle of a tool or a weapon, decorated with two lines of Aramaic inscription (Figures 3a, 4c, and 5) has recently been studied using an archaeometallurgical approach. It was found covered in a patina and corrosion products, and one side of the tube was more rounded than the other (the flatter side of the tube had perhaps been broken during the general use of the object). On the back of the tube, one side of the bent plate is folded over the top of the other side (creating an overlap between the two sides of the tube). The Aramaic inscribed letters (Figure 5a–c), varying in width between 200 and 1000 µm, were engraved using a sharp tool with a higher hardness than that of the copper-alloy tube. The tube was first cast into an open mould, perhaps made of sand, clay, or stone, to create a flat rectangular plate. The plate, between 2 and 5 mm thick, was then bent into the shape of a tube.

A small hole, 3 mm in diameter, based on its round shape and the area around it, was drilled close to the edge of one side of the tube, between the two lines of letters (Figure 4c, right side and Figure 6a). Logically, it is simpler to engrave the letters on a flat plate and only then to fashion it into its final tubular shape. However, as no deformation of the letters was observed, there is a high probability that the letters were engraved following the final shaping of the item. A bright, shiny orange metal was exposed after locally sanding the back of the object before performing the XRF analysis (Figure 6b).

As the current examined archaeological copper-alloy tube with an Aramaic inscription is a unique item, no destructive testing was allowed. HH-XRF chemical analysis of the external surface of the incised Aramaic tube patina layer revealed it to comprise 74.4–80.2 wt. % Cu, 0.7–0.9 wt. % Sn, 17.9–21.9 wt. % Si, 0.9–1.0 wt. % Fe, and up to 1.5 wt. % Al, as well up to 0.3 wt. % of the elements As, Zn, and P (Table 1). Preliminary XRF analysis of the patina layer performed by Lewis et al. (in press) also revealed a minor content of lead, as well as presence of trace elements such as Br, K, Mn, Ti, and V [1].

HH-XRF chemical analysis of the sanded surface indicated that the core metal was made of relatively high-purity copper, with composition between 89.4 and 96.4 wt. % Cu, 0.7–0.9 wt. % Sn, 2.0–8.9 wt. % Si, and 0.4–0.7 wt. % Fe, as well up to 0.3 wt. % of the elements As, Zn, P, and S (Table 1). During antiquity, artefacts made of relatively pure copper were rare, whereas items created using low-tin bronze (less than 1.0 wt. % Sn) were more common. Bronzes with different concentrations of tin were made by adding tin metal to the molten copper during the casting process [37].
Figure 5. The external surface of the copper-alloy tube with Aramaic inscription, showing the engraved letters covered in a patina (multifocal LM): (a) two engraved letters surrounded by bright green-yellow, dark green, white and black minerals; (b) engraved letter surrounded by brown-red, bright and dark green, white and black minerals; and (c) two engraved letters surrounded by bright and dark green, brown-red, white and black minerals.
Figure 6. The copper-alloy tube multifocal LM observation, showing (a) the small drilled hole surrounded by a patina cover and some corrosion products on the back of the object and (b) the bright metal areas that were locally sanded (back of the object) before performing the XRF analysis of the core metal; the dark areas are the remains of oxides and corrosion products.

Table 1. Results of the XRF analysis of the Aramaic-inscribed copper-alloy object.

<table>
<thead>
<tr>
<th>Measured Item Description</th>
<th>Composition (wt. %)</th>
<th>Other Elements</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Cu</td>
<td>Sn</td>
</tr>
<tr>
<td>Patina cover, area 1 (no sanding)</td>
<td>74.4</td>
<td>0.8</td>
</tr>
<tr>
<td>Patina cover, area 2 (no sanding)</td>
<td>76.7</td>
<td>0.9</td>
</tr>
<tr>
<td>Patina cover, area 3 (no sanding)</td>
<td>80.2</td>
<td>0.7</td>
</tr>
<tr>
<td>Metal, area 1 (locally sanded)</td>
<td>90.3</td>
<td>0.9</td>
</tr>
<tr>
<td>Metal, area 2 (locally sanded)</td>
<td>91.0</td>
<td>0.9</td>
</tr>
<tr>
<td>Metal, area 3 (locally sanded)</td>
<td>93.6</td>
<td>0.8</td>
</tr>
<tr>
<td>Metal, area 4 (locally sanded)</td>
<td>96.4</td>
<td>0.9</td>
</tr>
<tr>
<td>Metal, area 5 (locally sanded)</td>
<td>89.4</td>
<td>0.7</td>
</tr>
<tr>
<td>Metal, area 6 (locally sanded)</td>
<td>89.7</td>
<td>0.7</td>
</tr>
<tr>
<td>Metal, area 7 (locally sanded)</td>
<td>95.4</td>
<td>0.8</td>
</tr>
<tr>
<td>Metal, area 8 (locally sanded)</td>
<td>93.4</td>
<td>0.8</td>
</tr>
</tbody>
</table>

Based on the low tin concentration in the object’s alloy and the fact that the core base metal was made of relatively high-purity copper, its manufacturing process involved bending the artefact while heating and then fashioning the part into its final shape by
means of a small number of forging and annealing cycles. Consequently, the microstructure of the bulk alloy most probably includes recrystallized equiaxed grains and the presence of some slip bands, as well as annealing twins resulting from the applied thermomechanical processing. However, this assumption remains to be examined through metallographic analysis [26,29].

When the HH-XRF results obtained from the surface analysis were compared to the elemental values of the core metal after sanding the metal surface, a higher amount of copper and lower amount of the elements Si, Fe, and Al could be observed in the core metal (Table 1). The presence of Si, Fe, and Al is thus related to the corrosion processes and soil contamination [22]. The presence of 17.9–21.9 wt. % silicon in the external surface is thus probably related to the penetration of sand contaminants into the copper metal object through micro-cracks and many years of burial in an aggressive environment. However, there is also a possibility that the silicon was intentionally added to the copper alloy in order to improve the fluidity of the molten metal during the casting process, as well as to increase the strength and corrosion-resistance of the metal [38,39]. The positive correlation between the elements Si, Fe, and Al is indicative of the effect of environmental contamination.

Multifocal LM observation of the copper-alloy tube’s external surface before it was sanded revealed several colours of minerals, including bright green-yellow, dark green, white, black, and brown-red. The green colour may be attributed to atacamite, Cu₂Cl(OH)₃, and/or brochantite, Cu₄SO₄(OH)₆; the black colour to tenorite, CuO; and the brown-red colour to cuprite, Cu₂O [36,40].

Copper objects were designed and manufactured within a specific cultural sphere in antiquity. The use of copper and its alloys in ancient cultures depended on various factors, among them the ability to obtain raw materials (locally produced or via trade relations), the available metallurgical knowledge (and the know-how of existing manufacturing techniques), as well as the intended uses of the final copper object [23,26,31,32]. With the exception of silver coins and jewellery of the Persian period Levant, rarely has the metallurgical composition of epigraphic finds been chemically analysed. The current study establishes the evolution of our epigraphic object’s manufacturing processes while trying to provide an understanding of the technological process of copper production and its alloy in the Achaemenid Levant.

5. Conclusions

The object seems to have originally been part of either the handle or socket of a larger cultic vessel. The inscription, incised during its production (below), indeed indicates that it was not only used as a cultic object, but rather was produced for the purpose of cultic offerings, probably in a nearby workshop given the larger assemblage of inscribed copper-alloy artefacts from the site. All of these finds together provide evidence regarding the cultic nature of the site, possibly a roadside temple (and not just a favissa), located on the top of a high hill overlooking and controlling the area of a route that led eastwards from the central coastal plain to the Province of Samaria and its capital city, Samaria. The use of Phoenician and Aramaic script on the bowls and cymbal found at the site, and their varied ethnic connotations reflected in the personal names of their dedicators [12], as well as the recovery of Sidonian, Tyrian, and Samarian silver coins [13], may suggest a multicultural place of assembly in the 5th–4th century BCE Sharon plain.

The metallurgical analysis revealed that the core metal was made of relatively pure copper (89.4–96.4 wt. % Cu), with only a small amount of tin (0.7–0.9 wt. % Sn). The choice of a low-tin copper-alloy to produce the item indicates that the alloy was chosen based on technological considerations, in order to make it easy to bend and fashion the part into its cylindrical shape by means of plastic deformation. The manufacturing process of the object involved bending it while it was hot and then shaping it using several cycles of forging and annealing. Although the current metallurgical analysis has revealed the bulk composition of the object and the general manufacturing process, the microstructure of the core
alloy could not be observed because destructive testing was not allowed. In addition, the scope of the present study was insufficient to enable sourcing the origins of the raw materials. Future work, such as drilling metal from the bulk of the object and performing lead isotope analysis (LIA), may help to identify its provenance. However, because of the low amount of tin, it is possible that the object was produced from recycled copper, and if this is so, it will probably not be possible to determine the origins of the raw materials used to produce this object. The current study adds further information to the metallurgical knowledge and material culture of the Persian period Levant.

**Author Contributions:** The study was conceptualized by O.T. and D.A. All authors defined the research aims and contributed to the development of the research methodology. R.Y.L., E.E., and O.T. wrote the introduction section. D.A. wrote the metallurgical background and the experimental methods and tests sections. D.A. conducted the formal analysis. D.A. and O.T. wrote the original draft. O.T. was in charge of the project administration. All authors discussed the results and contributed to the conclusions. All authors have read and agreed to the published version of the manuscript.

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