

HIGHLY PROCESSED **BRONZE** JEWELRY AND ORNAMENTS IN THE **VÉRTESSZŐLŐS** FIND ENSEMBLE

ABSTRACT

Vértesszőlős was a populated area during the broad period of the Bronze Age. Artefacts from different cultures, and among them many bronze objects were found. Most are ornaments, but tools and weapons have also been unearthed. Jewellery and dress ornaments are found in many areas, and the examination provides an opportunity to compare regions, ages, and customs. Several stud ornaments and sporadic stud fragments were found during the excavation. Making studs requires careful elaboration and production. In general, it was stated that they were made of an alloy with a low and average tin content (3.5 -6 w% tin), which is malleable and easy to deform plastically. The morphology of the inclusions also suggests a high degree of plastic deformation. Due to the thin bronze material, the objects are subjected to solid corrosion effects, necessitating a detailed microstructural examination to examine the other constituents. The spiral beads require similarly careful elaboration, two of which were selected for parallel examination. A similar test protocol had to be used to determine the effect of corrosion. A slightly higher tin content and a more minor total deformation were found. The objects are also compared for inclusions and other impurities.

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INTRODUCTION

The Vértesszőlős site is in the northern part of Transdanubia, between Tata and Tatabánya, at the western foot of the Gerecse Mountains, on the bank of the Által stream, which curves towards the Danube (Fig. 1). Due to the stream valley leading to the river crossing, it is a geographically strategically crucial area that was inhabited in most archaeological periods. Therefore, the Makó-Kosihy-Čaka culture (Early Bronze Age), the people of the Encrusted Pottery culture (Middle Bronze Age), and the Tumulus and Urnfield cultures (Late Bronze Age) also used the area. Vértesszőlős was linked to Palaeolithic research, with archaeological excavations beginning in 1962, and the work related to the operation of the open-air site also led to the discovery of several new archaeological sites. For Bronze Age research, Vértesszőlős is primarily known for its Middle Bronze Age cemetery. Thanks to the results of excavations and site visits that continued with smaller or larger interruptions since 2005, new site and cemetery details, as well as bronze objects and a hoard find, enrich the Bronze Age material of the area (Vadász, Vékony 1979; Cseh 1999; Pál, Cseh 2013).

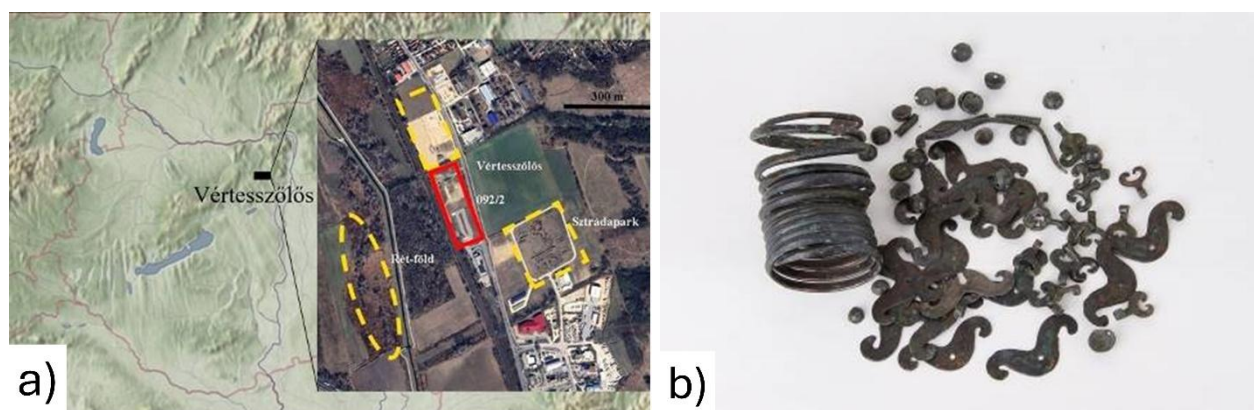


Fig. 1: The location of the Vértesszőlős site (a) and the hoard of Vértesszőlős (b).

Under the directorship of Gabriella A. Pál, excavations were conducted in 2009, directly southwest of the village, in the area between main road No. 1 and the Budapest-Győr-Vienna railway line, on the second terrace of the Által stream (Cseh 1999). These brought to the surface details of the Tumulus culture cemetery beginning in the Kosider period, a symbolic tomb or sacrificial ensemble from the Middle Bronze Age, as well as numerous scattered jewellery and fragments of a dagger unearthed because of humus digging and inspection work. Of the eight graves excavated in the cemetery, three were cremations, one symbolic, and the rest were inhumation burials. All the latter tombs were disturbed, producing bronze pins and eared mugs. In addition to the pins with curved stems and nail heads, the appendages of the women's grave were two pierced bronze discs, presumably hanging from the belt, two disc-headed pins (one decorated), and further bronze pins adorning the dress. In addition to/among the known types of jewellery of the Encrusted Pottery culture (swallowtail or Ω -shaped pendants) (Szabó 2022), a so-called 'anthropomorphic' pendant was also found among the scattered material, along with spiral beads, bronze studs and a fragment of a bronze dagger.

The excavation started with removing the topsoil of the entire development area to a depth of 20–40 cm. Both the removed topsoil and the mixed surface between the objects were investigated with metal detectors, thanks to which numerous metal objects, mainly from

Roman times, were found. The result of the previous excavation was about 37 objects from the Bronze Age, including settlement material of the Encrusted Pottery culture, the barrows of the early Tumulus culture, as well as details of Roman and Árpád period settlements (Cseh 1999). Lajos Sándor found the bronze hoard only after the excavation was completed, on the agricultural land on the north side of the cuttings, where the Encrusted Pottery and Tumulus culture cemeteries also continue, during metal detector searches. The hoard came into the possession of the museum of Tatabánya in 2016 as a gift (Fig. 1).

The metallographic examination of metal objects provides information about the alloy and state of the object, so the main steps of the manufacturing technique can be determined (Török *et al.* 2013;. 2017a). The mechanical properties of objects can be estimated based on microstructure analysis, even if it is impossible to examine them (Barkóczy *et al.* 2012; Gyucha *et al.* 2015). Of course, this requires knowledge of the characteristics of each state: cast, plastically deformed, and annealed alloys (Póliska *et al.* 2006; Barkóczy *et al.* 2003). Evidently, it can also deduce the critical parameters of the processing of the object from its shape and usage, so information can be obtained even if only the composition can be examined (Benkő, Barkóczy 2017). The alloys and manufacturing techniques reveal specific features of the given metalwork object, supporting archaeological analysis (Horváth *et al.* 2020; Török *et al.* 2017b). Studies focus primarily on high-importance, specialized objects such as weapons (Sureda *et al.* 2021; Mödlinger *et al.* 2013). Several types of objects are examined when analysing hoard finds and collections (Mödlinger, Trebsche 2020; Oudbashi, Davami 2014). Metallographic examination is a destructive material-testing procedure where objects must be sampled, and samples prepared for examination by mechanical and chemical treatment (Scott, Schwab 2019). When taking samples, the sample itself must be characteristic of the object. However, heritage protection principles and restoration methods must also be considered (Szabó 2010; Kovács *et al.* 2021).

This article presents the analytical results from the study of the hoard find and scattered studs of Vértesszőllős. The production of studs requires a large amount of processing of the metal as raw material, which suggests a unique manufacturing technique. Therefore, the results from the analysis of these samples provide an excellent example of the characterization of metalworking techniques. In addition, the artefacts also contain spiral beads with similar processing requirements, from which two objects were sampled for examination. Thus, comparing the two types of objects provides additional information for the analysis of metalworking.

MATERIALS AND METHODS

The test programme examined samples of six studs and two spiral beads. As the objects are fragmented, a small sample was taken of the fragmented part, in order to leave the nature and appearance of the object unaffected. This means small samples, all of which were embedded in a two-component resin for the examinations. Then, the resin surface was machined, and the metallic surfaces were prepared for metallographic examination. The samples were polished with SiC abrasive grains and diamond paste with 3 and 1 µm grain sizes. Both grinding and polishing were complicated because, due to the nature of the objects, the metallic surface to be prepared is small compared to the corrosion layer on the surface of the objects. Particles peeling off from the corrosion layer often scratched the surface. After careful mechanical preparation, the samples were etched in an aqueous solution of K₂CrO₄ by dipping method. Due to the potent oxidizing agent, the oxide layer formed on the surface makes the grains colourful under polarized illumination. However, in a

bright field illumination, the morphology and structure of the inclusions can also be examined. Optical microscopic observations and images were made with a Zeiss AxioImager M1m microscope. After optical microscopy, the samples were analysed using SEM-EDS method without further preparation on a Hitachi 4300 CFE. The sample's average chemical composition and, occasionally, the phases' local analysis were determined during the measurements. When examining phases, element maps were used instead of compositional data, because the main goal was identifying phases instead of accurate chemical analysis. From the results, the composition of the raw material was determined, and the characteristics of the manufacturing technique were established. From the examination analysis of the individual objects, we can deduce the specific characteristics of the artefact.

RESULTS

Optical microscopy of the 092-2/1 stud reveals a medium-sized recrystallized grain structure (Fig. 2a). This suggests that the material of the stud was annealed after plastic deformation. Highly elongated inclusions are found in the microstructure (Fig. 2b). The inclusions are copper sulphide based on the SEM-EDS analysis (Fig. 2c). Copper sulphide is a soft, malleable phase, so that when the material is plastically deformed, it elongates in the direction of the material flow while thinning perpendicular to it. The elongation of the phases suggests that the material of the stud has undergone considerable plastic deformation, which is also shown by the shape and nature of the object.

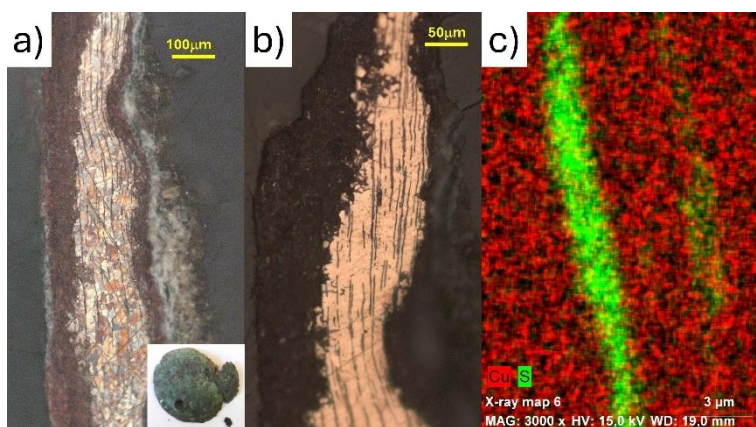


Fig. 2: The microstructure of the 092-2/1 foreskin in polarized illumination (a). A recrystallized grain structure can be seen. In a bright field illumination, largely elongated inclusions can be seen (b). The inclusions based on the chemical composition analysis are copper sulphide inclusions (c).

The average chemical composition of the stud was determined by SEM-EDS measurement in an area visibly not affected by corrosion. In addition to copper, the alloy contains 6.14 wt% tin and 1 wt% arsenic (Tab. 1). Of course, it also contains sulphur due to sulphide inclusions, but the volume ratio of the phases is such that the average composition analysis did not detect the sulphur content. Mechanical properties are more affected by tin, due to its quantity. This tin content gives good deformation properties when plastically deformed, but the alloy remains sufficiently hard. Under the influence of cold plastic deformation, the alloy hardens, increasing with the degree of deformation, which means a more significant force of further forming. In this case, the hardness can be reduced by annealing. Recrystallization occurs in the plastically deformed microstructure, which can also be seen in the grain structure. The degree of deformation was probably high before annealing, and softening could occur at medium temperatures. In this alloy, annealing occurs above 300°C in a

relatively short time. The softening was crucial, because it was possibly the last manufacturing phase before finishing the surface of the stud.

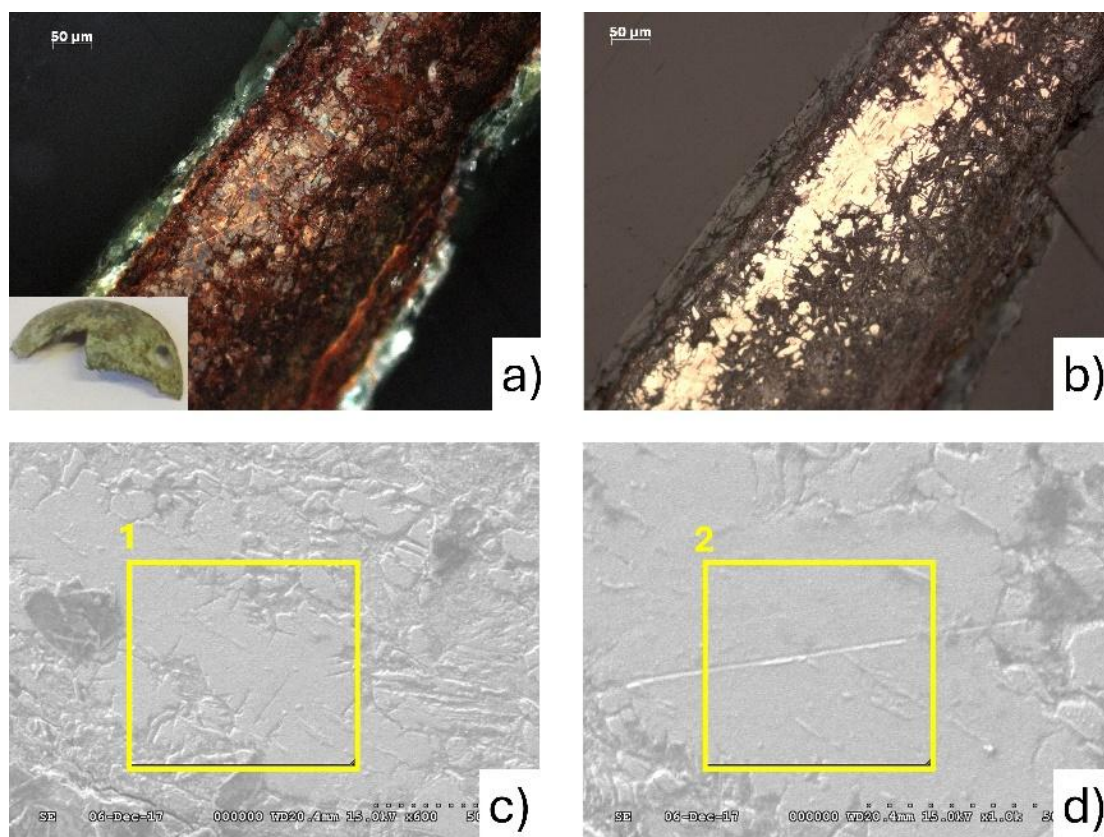


Fig. 3: Micrographs of the microstructure of the 092-2/2 stud. The sample is heavily affected by corrosion. In polarized illumination, a recrystallized grain structure can be seen (a). In a bright field illumination, elongated sulphide inclusions of small size are observed (b). SEM-EDS analysis was performed in several areas close to (c) and farther (d) from the corrosion layer as far as the metallic volume of the sample allowed.

A wide corrosion layer can be observed in the sample from the 092-2/2 stud, with little residual metallic volume (Fig. 3). This makes object analysis difficult, especially the chemical composition analysis (Tab. 1). A significant amount of oxygen was measured in each analysed area (Fig. 3c and 3d), indicating the presence of copper oxide. However, the EDS analysis gives the oxygen content with great uncertainty rather than as indicative data. In our case, we can still get enough results for comparison next to it. It is heavily burdened with corrosion (Fig. 3c). A relatively high tin content of 9.8 wt% can be measured, but in the area less exposed to corrosion (Fig. 3d), only 2.3 wt% tin can be measured. The segregation of tin in the corrosion layer due to corrosion processes is a known phenomenon that can be significant. Therefore, the measurements must be sufficiently accurate regarding the tin content. All that can be stated is that the alloy contains tin, but its amount is around 2 wt%. In addition to tin, a little silver is detected in the alloy. Arsenic can only be detected in small amounts in the corrosion product, which is also due to segregation. The alloy indeed contains detectable quantities of silver and arsenic (Tab. 1). Low sulphur content can also

be measured in the corrosion area, due to copper sulphide inclusions visible in the microstructure (Fig. 3b).

The low tin content and the other components mean an alloy that is highly malleable when cold, from which the thin material of the stud can be easily formed. Of course, hardening is also characteristic of this alloy, so after considerable deformation, it must be annealed to facilitate further deformation. In this case, annealing occurs before finishing, resulting in the recrystallized grain structure. The elongation of the well-malleable copper sulphide inclusions also indicates a high degree of plastic deformation. In this sample, the elongation of the inclusions is not as significant as in the alloy of the previously examined stud. However, it must be remembered that the direction of deformation cannot be determined from the stud's appearance, and the sampling place was fixed. Therefore, we cannot deduce the specific degree of total deformation from the elongation of sulphide inclusions. It can be stated that the material of the stud was subjected to a large amount of complete deformation. However, this finding is sufficient for comparison.

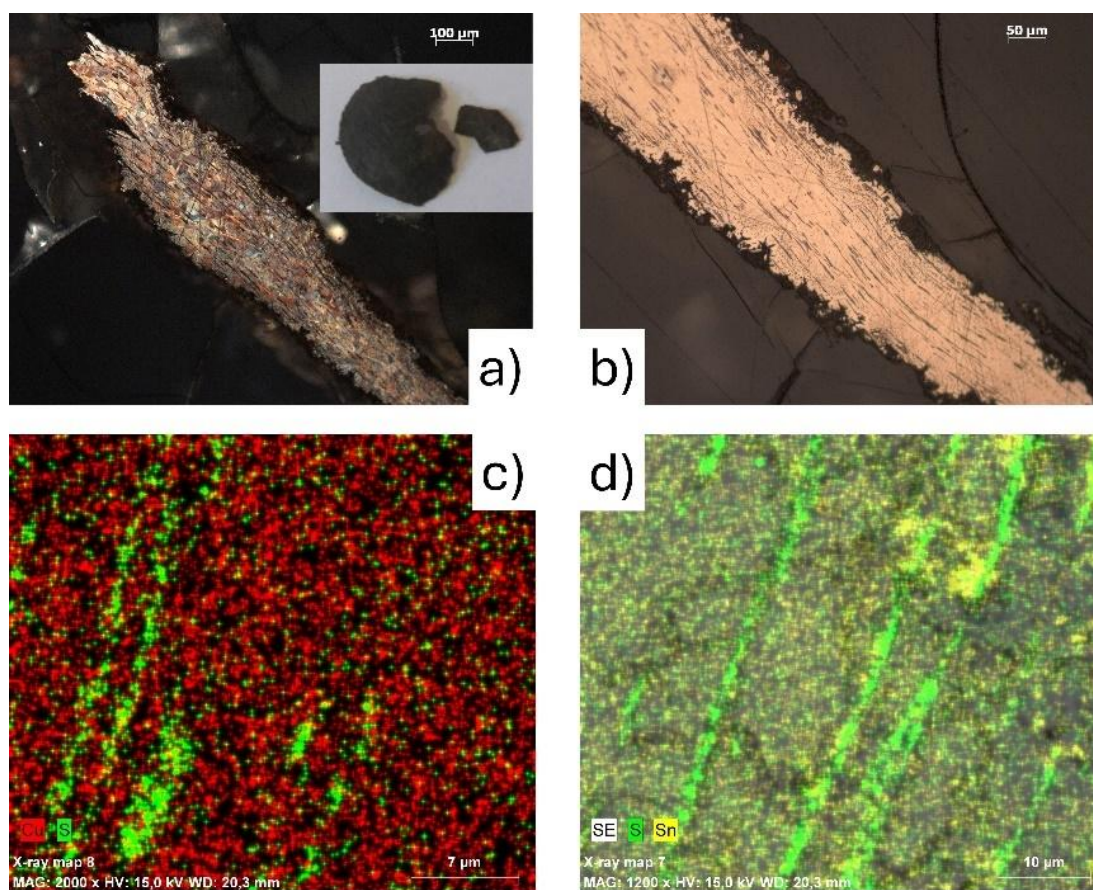


Fig. 4: Micrographs of the microstructure of the 092-2/3 stud. In polarized illumination, a recrystallized grain structure can be seen (a). Elongated inclusions are observed in a bright field illumination (b). SEM-EDS analysis showed that copper sulphide inclusions are the elongated phases in the microstructure (c, d).

The 092-2/3 stud is lightly burdened by corrosion, so that the metallic volume can be well examined. In this case, optical microscopy also revealed a recrystallized grain structure (Fig. 4a) and highly elongated inclusions (Fig. 4b). The elongated inclusions are also copper sulphide inclusions (Fig. 4c and 4d). The average composition of the sample is high in tin at

11.6 wt% (Tab. 1). This tin content already means high hardness and strong strain hardening due to cold forming. In addition to tin, arsenic, and sulphur can be detected in small quantities (Tab. 1). Still, they have no significant effect on the behaviour of the alloy in terms of measured amounts.

As in the analysis conducted as part of previous studies, the strong elongation of sulphide inclusions supposes a significant degree of complete plastic deformation. Such a high tin content means intense strain hardening, so even more annealing may have been required during the manufacturing. However, as can be deduced from the recrystallized grain structure, annealing also occurred before finishing. The grain-structure characteristics assume heat treatment conditions similar to the previous examinations. From this point of view, similar manufacturing must be considered, based on the high degree of strain hardening caused by the tin content.

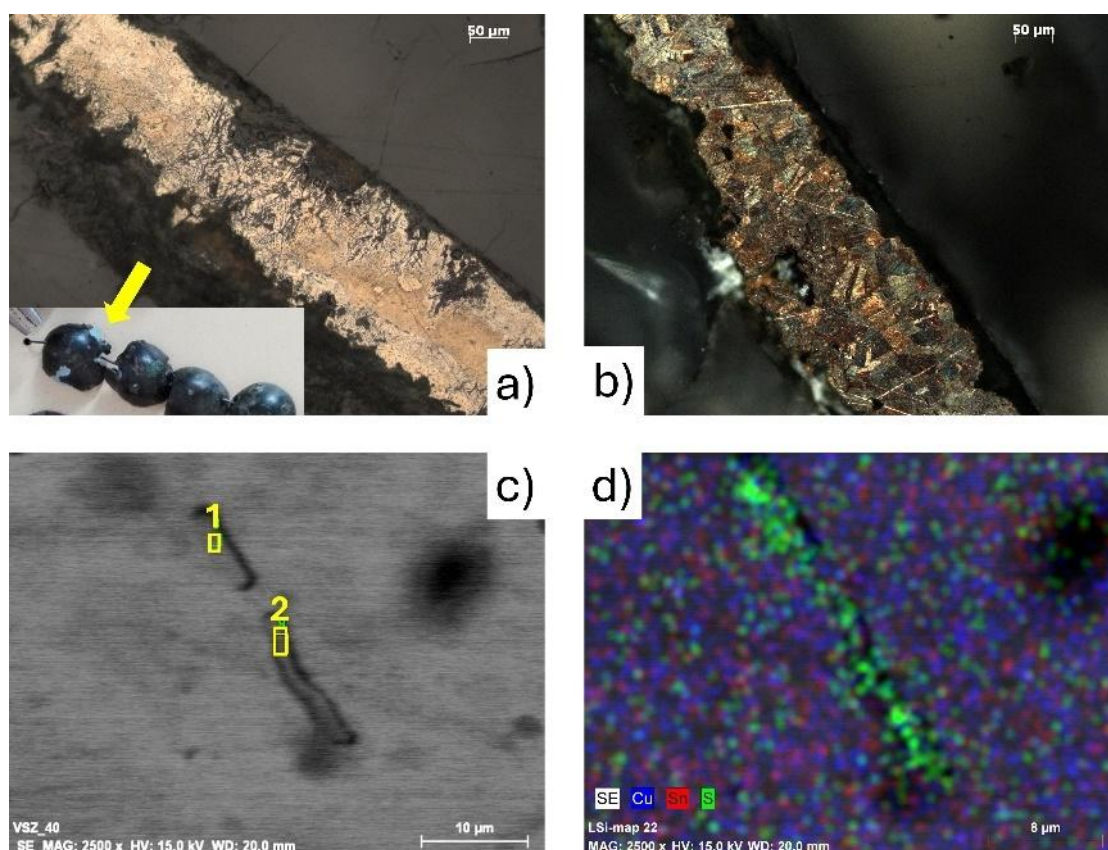


Fig 5: Micrographs of the VSz40 stud. The material of the stud is heavily affected by corrosion (a). However, the metallic volume can be analysed. Polarized illumination shows a recrystallized structure of medium grain size (b). Small-sized, elongated inclusions in the microstructure are found in relatively tiny amounts (c). EDS analysis identifies these inclusions as copper sulphide inclusions (d).

Studies so far have shown three studs made similarly from different alloys. VSz40 is challenging to analyse, due to strong corrosion effects, but the metallic volume is sufficient to ensure proper results (Fig. 5a). The microstructure of the stud consists of recrystallized grains (Fig. 5b). A relatively tiny amount of small-sized elongated inclusions can be found in

the microstructure (Fig. 5c). EDS analysis showed that copper sulphide inclusions were found.

The chemical composition analysis revealed a moderate tin content (5.7 wt%) (Tab. 1). In addition, little arsenic can still be detected in the alloy. From this point of view, the material of the stud resembles the alloy of the first tested stud, 092-2/1. Iron and nickel can also be detected in different amounts in the inclusions, but their quantity is not decisive for the composition of the alloy (Tab. 1). The morphology of both the alloy and the inclusions assumes a similar manufacturing technique as in the case of the previously studied 092-2/1 stud. Of course, this similarity in the production technique is not surprising considering the nature of the objects, since their thickness, shape, and appearance are almost identical. However, differences in their composition are more straightforward to analyse in the light of manufacturing.

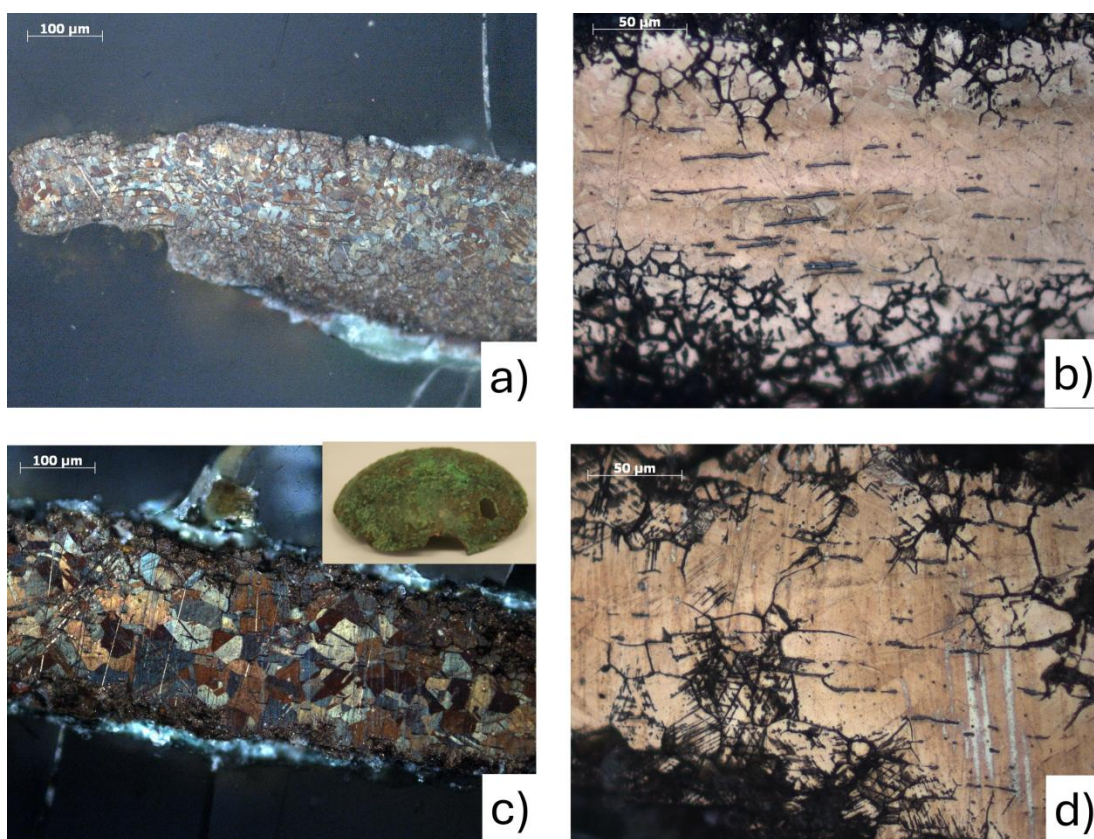


Fig. 6: Microstructure of P14 and P26 studs. The recrystallized grain structure of the P14 stud is shown by (a). In the microstructure, elongated sulphide inclusions are seen (b). The P26 stud microstructure also consists of recrystallized grains (c). However, the grain size is significantly larger. In its microstructure, fewer but also elongated sulphide inclusions can be seen (d).

The microstructure of the P14 stud is similar to that of the other studs examined (Figs 6a, 6b). Recrystallized grain structure and large, elongated sulphide inclusions were revealed. In terms of chemical composition, it contains little tin (3.5 wt%) (Tab. 1). However, in addition to tin, a significant amount of arsenic can also be measured (2.2 wt%), which already affects the properties of the alloy. The amount of silver and antimony of around 1 wt% should also

be highlighted, distinguishing the stud's material from other alloys. In addition, lead and large amounts of bismuth can be measured (Tab. 1). Similar statements can be made about its manufacturing as in the case of other tested studs.

Analysis of the microstructure of the P26 stud also revealed a recrystallized grain structure (Fig 6c). However, the grain size in this stud is significantly larger. This difference can be found in the conditions of annealing. In addition to the moderate tin content (5.5 wt%), iron, nickel, silver, and arsenic can be measured (Tab. 1). The lead content is highest in this sample but is also considered contaminating, like the other components mentioned alongside tin. Few but elongated copper sulphide inclusions also show a high degree of total deformation, which justifies the assumption of annealing. The larger particles show longer, high-temperature annealing in the case of this stud, during which, in addition to recrystallization, grain coarsening also took place. However, the basic scheme of manufacturing is identical to that established earlier.

Object	Fig.	S	Fe	Ni	Cu	As	Ag	Sn	Sb	Pb	Bi	Rem.
092-2/1	Fig. 2, average				89,37	0,99		6,14				O, C
	Fig. 3, average											
092-2/2	(1)	0,32			23,86	0,25	0,53	9,78				O, C
	Fig. 3, average											
	(2)				11,02		0,14	2,27				O, C
092-2/3	Fig. 4, average	0,62			68,77	0,06		11,58				O, C
	Fig. 5, average				84,63	0,42		5,67				O, C
VSz40	Fig. 5, phase											
	(1)	8,37	0,42	0,05	66,81			3,98				O, C
	Fig. 5, phase											
	(2)	8,84	1,37	0,29	63,76	0,33		2,31				O, C
	Fig. 6.b,											
P14	average	0,1			87,59	2,24	0,99	3,5	0,94	0,61	2,08	O, C
	Fig. 6.d,											
P26	average		1,19	1,42	84,83	2	0,22	5,54		1,45		O, C
SCS	Fig. 7, average			1,04	87,79	0,03		6,91	0,52	0,4	0,23	O, C

Tab. 1: The results of the EDS analysis. The concentrations of different elements are in wt%.

The Vértesszőllős artefact assemblage also contains spiral beads, of which two could be examined. Spiral beads were chosen because they have similar processing needs. However, it is necessary to make them into a wire-like form, which is wound up later during the manufacturing of the object. For this reason, even the size of the objects does not allow large samples to be taken in this case. The microstructure of the SCS spiral bead is shown in Fig. 7. The recrystallized grain structure (Fig. 7a) also refers to annealing before finishing in this type of object. Here, the final manufacturing operation is presumably the formation of the spiral, which causes a slight residual deformation that is not detectable in the grain structure. However, a high degree of deformation is suggested by the elongated copper sulphide inclusions (Fig. 7c and 7d). Strain hardening makes the deformation of the spiral significantly more difficult. For this reason, annealing was necessary to eliminate the effect of hardening and to allow the formation of the spiral to be carried out.

The spiral bead contains a medium amount of tin, 6.9 wt%. In addition, the nickel and antimony content should be highlighted from the point of view of the alloy (Tab. 1). Low lead and bismuth content can also be detected. Of course, here, too, the hardness of the alloy and the degree of its strain hardening are primarily determined by the tin content. Here again, it can be said that this alloy is very malleable in cold forming. However, due to the intensive strain hardening, the large amount of deformation that can be assumed based on the elongation of the sulphide inclusions necessitated annealing, as stated above. Even a few tiny compact phases can be observed in the microstructure. EDS analysis revealed significant levels of tin and antimony. This suggests that these phases are intermetallic phases of a tin-antimony-copper alloy system. They probably have been formed during the alloy's crystallization due to microsegregation.

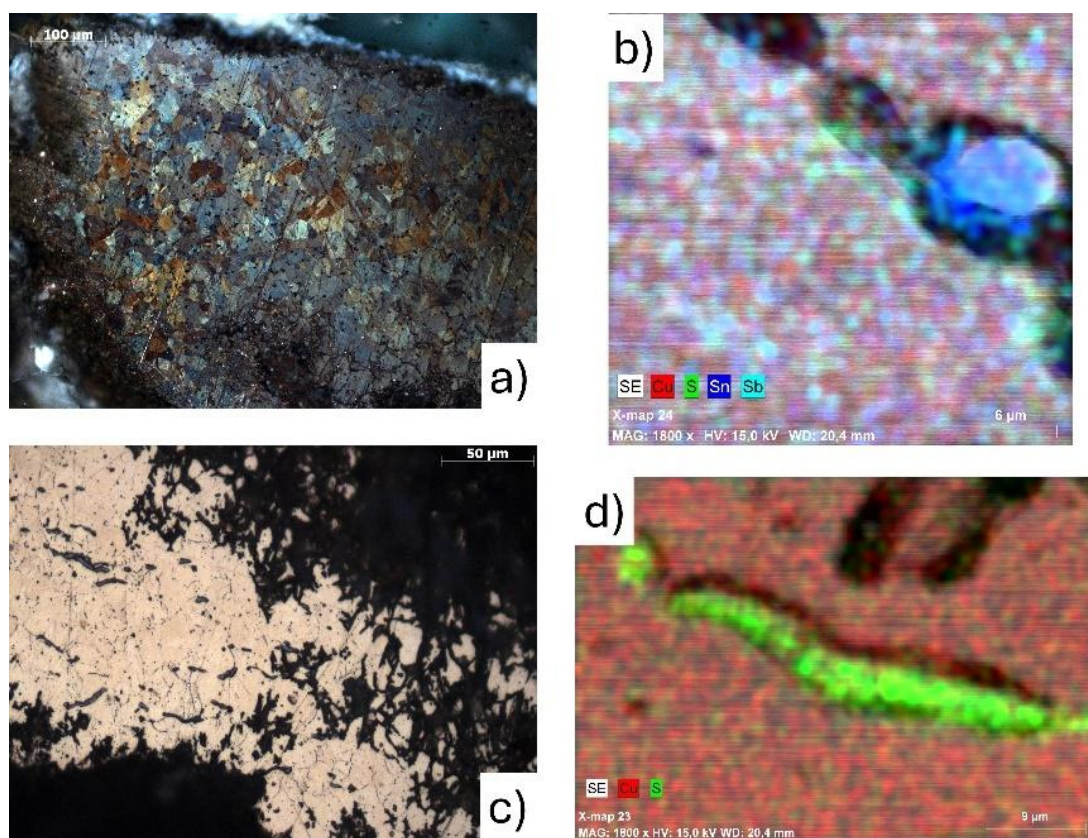


Fig. 7: Microscopic images of the SCS spiral bead. Polarized illumination shows a recrystallized grain structure (a). Large elongated and small spherical inclusions are observed in the structure (c). The more prominent elongated inclusions are sulphides (d). Tiny spherical inclusions are intermetallic phases (b).

Similar observations can be made about the microstructure of the spiral bead VM, except that there are few inclusions in the microstructure. The sample was taken from the spiral cross-section, so the inclusions' elongation must also give an adequate picture of the proper degree of total deformation. The average composition analysis revealed a relatively high tin content (8.67 wt%) of copper at 83.69 wt%. In addition, low levels of arsenic (0.2 wt%) and silver (0.47 wt%) were detected. This composition is similar to the second 092-2/2 stud tested. Regarding production technique, similar statements can be made.

CONCLUSION

Studs and spiral beads are all processing-intensive objects that require a great degree of deformation. In the case of both object types, the significant plastic deformation of the base material causes high-strain hardening, which makes the last production phases especially difficult to perform. For this reason, annealing was required at least before this step. During annealing, the recrystallization process eliminates strain hardening, and the alloy becomes easier to deform again. For each object, the recrystallized grain structure was detected. The shape of malleable copper sulphide inclusions indicates a high degree of deformation. In most cases, greatly elongated inclusions can be seen. Copper sulphides elongate in the direction of material flow during plastic deformation. However, this elongated shape does not change during annealing heat treatment, so the degree of all deformations to the alloy can be estimated from this. Of course, this is only true if we can examine a sample that contains the direction of material flow. In this case, sampling was fixed, so it could only be determined that the deformation was so large that softening was necessary before the last step of manufacturing. It is also only possible to decide whether this one or more annealing has been carried out in the preparation process. However, the degree of strain hardening is revealed by the composition. It is mainly the tin content that determines the mechanical behaviour of alloys in the case of the objects under consideration. It is generally known that the higher the tin content, the more intense the strain hardening.

The similarity of the manufacturing technique between the objects is not surprising, since the nature of the objects determines the same main steps and possibilities of production. Only the parameters of the identified annealing treatment can differ in the case of particular objects, which can be seen through the size of the recrystallized grain size. This also includes the degree of current plastic deformation of the annealed metal, so it cannot be projected on its own, only at temperature or during heat treatment. However, considering this, the test results revealed a similar manufacturing procedure.

However, there is a much greater variety in the alloys used. The tin content itself also varies, from a low to a relatively high tin content. Bronzes were also found when examining the raw materials of the objects. However, what is even more interesting is the presence and quantity of other elements. Iron, nickel, arsenic, silver, and antimony are highlighted. The amount of these elements is related to the composition of the base copper, the composition of the ore, and the conditions under which it was smelted. Previous studies have identified alloy types into which the raw materials of objects from the Carpathian Basin can be classified. It is possible to find almost all of the alloys of studs and spiral beads. One important group is the alloy containing arsenic and silver, which includes the 092-2/2 stud and the VM spiral bead. The other significant group contains iron, nickel, arsenic, and nickel. Silver is often present only to such a small extent that it can only be detected when segregated in the corrosion product. This group includes VSz40 and P26. However, only arsenic was found in the alloy in addition to tin. The presence of antimony creates separate groups. Of course, in the case of antimony, it is a question of whether it enters the alloy as an impurity of copper or tin, so it is essential to examine the alloys in detail. In this case, the measured small quantities and alloys bind the presence of antimony to the copper base.

The result of the incredible variety in alloys is that similar products have been formed using similar techniques from raw materials from multiple sources. The significant array of tin content confirms this. This observation is consistent with previous studies that there was no stable supply source, but that smiths also used various available sources to make the

objects. The test results confirm this in the case of similar objects, where the variety of objects and differences in the production technique do not leave open questions.

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