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People and Donations
Over the last year, the renewed activity of IAM S has resulted in a number of stimulating and promising developments. Following the most generous donation by BHP Billiton plc of a three-year studentship in archaeo-metallurgy (see IAM S 21), we received several very strong applications from prospective students. IAM S’s Scientific Committee eventually decided to split the studentship between the two most outstanding candidates for the Institute’s M Phil / PhD programme. The main recipient is Xander Veldhuijzen, who has previously studied at Leiden University, where he co-excavated Tell Hammem az-Zarga, a multi-period site in northern Jordan, as part of a joint mission between the Yarmouk University of Jordan and Leiden University. The site turned out to contain remains of a significant smelting and refining activity for iron, probably relying on the nearby ore deposit of Mugharet al-Warda. The scale of smelting activity as demonstrated by the sheer volume of slag and the interesting nature of this slag clearly justify a closer study of this site, which is one of the earliest in the Middle East to yield evidence of iron smelting. An additional support will be given to Myrto Georgakopoulou, a student with a first degree from Imperial College now studying Early Bronze Age copper smelting in the Cyclades, with particular emphasis on the island of Seriphos.

It is the continuous generosity of the Sternberg Foundation that allows us to co-sponsor this year Shadreck Chirikure, a student from Zimbabwe, to do his master degree at the Institute. Shadreck is a typical example of a very bright student who, had he not have received substantial funding from a number of sources, could not possibly have even begun his studies here. For his dissertation, he plans to study iron smelting and mining in Nyanga, a region in south-eastern Zimbabwe, where no such study has yet been performed. I am particularly glad that IAM S can contribute to his funding; it highlights the wide international character of archaeo-metallurgy at the Institute of Archaeology, both among its students, and the research topics pursued.

In welcome addition to the ongoing institutional and corporate support, a number of new individuals from the modern metal industry have joined IAM S this year, demonstrating that they accept the responsibility of the current industry for its unique heritage. Their enthusiasm and engagement places them in the tradition of those mining engineers and metallurgists who, over the last hundred and fifty years or so, have recorded, studied and preserved the traces of activity of their forebears from times immemorial. It was - and is! - these individuals who laid the foundation for the much-younger academic discipline of archaeo-metallurgy, and we at IAM S are proud to serve at the interface of industry, trade and academia.

Timna
The New Timna Park is now open to the public. The first of two buildings of the Visitors’ Centre, dedicated to Egypt’s culture as background to the Egyptian copper industry in Timna, has been completed and is now open, also the new open air exhibition Rockart in the desert. Work on Phase Two is about to start and Phase Three is in the planning stage. Phase Two will consist of replicas of all periods of ancient mining in the Arabah, as a visit of the original mines would be too dangerous for the public and the mines as well. It is intended to be as close to the original as possible to enable the visitors to experience mining underground during the different periods. Some difficulties were experienced during the recent excavations in the mines. The assistant in charge of the excavations, Alexandra Sion, experienced difficulty in breathing whilst in the underground galleries. She felt that, as she was finding this a serious problem, it would also have been a problem for the ancient miners. With this in mind, she looked for an ancient solution to this problem in the workings – and discovered a number of ventilation shafts previously not identified. This discovery has stimulated further important research in the layout of these mines, now being conducted jointly with Tim Shaw.

Journal
This is the second issue of the IAM S journal in its new style. Its content mirrors the increased submission for publication of high quality papers from all over the world, from Israel, Greece, Romania and Britain to Argentina, covering several millennia of metallurgy. I am particularly grateful to the various referees who assisted me in the selection and editing of the papers for this issue, and their helpful suggestions to the authors for improvements. I very much encourage readers to consider submitting their own papers for publication in the IAM S journal. Any journal can only be as good as the papers offered to it for publication, and I hope for the continuous support of our readers and colleagues to further develop IAM S to a high-quality, fully refereed journal.

This has led to an increase in the number of pages in this issue over the previous ones; while this is certainly a good sign for the healthy future of our journal, it is also a matter of concern from a financial point of view. Please contact me if you would like to consider giving a charitable donation, however small or large, to the Institute for Archaeo-Metallurgical Studies in order to keep its journal as a free service to the metal community old and new.

Teaching
The teaching of archaeo-metallurgy at the Institute of Archaeology is well established throughout the programme of study, at 3rd year undergraduate level, within the Institute’s MSc programmes, and at MPhil / PhD level. Following the recent restructuring of the Institute’s teaching programmes and the establishment of the new Chair of Archaeological Materials and Technologies, the traditional MSc in Archaeo-metallurgy has grown into the new MSc in the Technology and Analysis of Archaeological Materials, currently in its second year. The back page of this issue has additional details of this degree, which covers all aspects of such inorganic archaeological materials as metals, glass, lithics and ceramics in their production, use and meaning in past societies. I am always happy to answer more specific questions regarding its content and structure, and application. Several of last year’s master dissertations which were finished only in mid-September have already resulted in publications being submitted to excavation monographs and journals, not least in this issue; several others are in preparation. This underlines the strong research tradition of both the Institute for Archaeo-Metallurgical Studies and the Institute of Archaeology, and the cross-fertilisation of teaching and research that we experience here so strongly.

In addition to the regular teaching programme for students enrolled at the Institute of Archaeology, we will offer two one-week Summer Schools this May. Brief details follow:

Professors Tim Shaw and Beno Rothenberg will present and discuss ancient mining technology, drawing on their rich own expertise in the field across the world. The second week, beginning on Tuesday 21 May, will provide an introduction to the principles of ancient metallurgy and the study of metallurgical remains. The lectures here will be predominantly by Professors H G Bachmann, Beno Rothenberg and Thilo Rehren. The fee for each week will be only GBP 50, and people potentially interested are invited to contact me via email (th.rehren@ucl.ac.uk) or mail (at the Institute's address) for further programme details, and registration.

Thilo Rehren
Matthew Ponting

Abstract
This paper presents the chemical analysis of copper-alloy metalwork from three early first century sites in the Galilee area of modern Israel together with their interpretation and discussion. The analyses were by Inductively Coupled Plasma Atomic Emission Spectrometry (ICP-AES). Interesting compositional differences were found between those artefacts from Jewish sites and those from a pagan site which mirror differences already identified in the material culture assemblages. Brass, an alloy with strongly Roman associations, was found to be an alloy only present in the pagan assemblage, whereas traditional tin-bronze dominates the assemblages from Jewish sites. It is suggested that, during this very polarized period, the populations of the Galilee declared their cultural affiliations through the manipulation of the materials they used as well as the objects they manufactured and traded. Parallel offering explanations for this phenomenon were found in other material culture studies and the reasons for these ‘choices’ are investigated.

Introduction
The adoption of brass and the mastery of the necessary technology needed to produce it, is one of the many changes and developments in material culture that accompany the expansion of Roman influence in Western Europe (Bayley 1998: 19; Dungworth 1997). During the first century BC and pre-conquest years of the first century AD, the local tribal elites in those areas peripheral to the Roman provinces were keen to acquire Mediterranean luxury items - prestige goods - to reinforce their status at home (Milliet 1990; Collis 1984: 158-180). And the appearance in the indigenous metalwork of peripheral peoples noted above, it was felt of value to ascertain whether the same phenomenon occurred in Palestine. If it did not, then copper alloy metalwork was probably subject to the same strictures as the other aspects of material culture identified by Berlin.

Methodology
The approach adopted was to analyse copper alloy artefacts from three excavated sites in the Galilee region (Fig. 1). The sites chosen were all significant settlements (towns rather than cities) during the period of this study and, furthermore, were substantially destroyed during or shortly after the revolt and not subsequently re-occupied (Syon 1993; Adan-Beyewitz & A via 1997; Herbert 1994). All the material analyses were as part of a separate project (Ponting forthcoming a), but provide a useful body of material that is unquestionably Roman. The number of copper alloy artefacts from each site is not large, numbering at best in the 20’s and 30’s, but in fact, the pieces that were in good enough condition to allow sampling was a fairly small subset. Consequently, all the objects in each excavated group, which were from appropriate contexts, and which were in a condition to warrant sampling, were sampled. Thus the sample assemblage is about as representative as the archaeology will allow.

A recent study of the material culture of the towns and cities of the Galilee region of Northern Israel has shown a quite different trend during the same period - the early years of the first century running up to the Great Revolt of A.D. 66 (Berlin 2002). Here it appears that the increasing antagonism towards the Romans by the Jewish population (as recorded in the writings of Flavius Josephus) is reflected in material culture by the increasing exclusion of objects with clear Roman associations in assemblages from Jewish settlements. Given the close association of brass with Roman material culture and its gradual appearance in the indigenous metalwork of peripheral peoples noted above, it was felt of value to ascertain whether the same phenomenon occurred in Palestine. If it did not, then copper alloy metalwork was probably subject to the same strictures as the other aspects of material culture identified by Berlin.

Fig. 1. Map of the Western Galilee area showing the location of the sites discussed in the text.
The analytical technique adopted for this project was Inductively Coupled Plasma Atomic Emission Spectrometry (ICP-AES). This technique is highly sensitive, measuring minute traces of contaminants down to fractions of a part per million, and so is ideal for investigating the contamination of local bronzes by brass or other zinc containing alloys. This sensitivity also allows for the study of other trace elements that are solely related to the geochemistry of the ores smelted and/or the smelting technology used. The unquestionable likelihood that the metals of these artefacts have been repeatedly recycled means that it would be hopeless to attempt to attribute any object to a particular source. Nevertheless, it is often possible to 'characterise' an alloy and ascribe it to a compositional group that may, or may not, correspond to an archaeologically useful feature, such as style or site location. The operating parameters of the ICP instruments used are presented in the reports on the individual assemblages (Ponting forthcoming b, c, d). Likewise, the analytical data for the artefacts from each site are presented in the appropriate reports.

Results and discussion

For all of the three settlements sampled, the majority of the artefacts (70.3 % overall – 52 pieces) were made from a moderate to low tin-bronze (mean of 6.4 % tin). U-n alloyed copper was used for 24.3 % (38 pieces) of the artefacts, whilst brass accounted for only 5.4 % (4 pieces).

Only one site, Tel Anafa, had any brass artefacts (zinc contents between 17 % and 21 %), whilst all three had objects of bronze and copper (Fig. 2). The brass objects from Tel Anafa are all decorative rather than utilitarian; two fibulae, a bracelet fragment and a cosmetic spatula (Ponting forthcoming c). At Yodfat, bronze and copper made up equal proportions of the twenty objects analysed (ten of each metal). Gamla showed the highest proportion of bronze (86.8 % - 33 pieces) with the remainder of un-alloyed copper. Tel Anafa and Gamla had broadly similar proportions of un-alloyed copper (18.8 % and 13.2 % respectively).

The results of the analyses therefore fit with the model proposed above. The only brass objects occur on the single pagan site, making up 25 % of the total assemblage. Whilst admittedly the sample numbers remain small, the samples do account for nearly all the available excavated metalwork from each site and so must be reasonably representative of the metalwork in use during the period. Additionally, if we see the use of brass as a companion to bronze rather than copper and largely reserved for decorative purposes, rather than utilitarian, the addition of percentages of brass and bronze at Tel Anafa (81.3 %) is very close to the percentage of bronze alone at Gamla (86.8 %). The situation at Yodfat is clearly different, with equal copper and bronze.

The brass items from Tel Anafa are all made of a generally good quality, high zinc cementation brass with little contamination by other elements, which is a trait shared with most first century Roman brasses. The single exception to this is the brass bracelet fragment, which contains a little over 1 % of tin and almost 1 % of lead. However, such small amounts can also be seen in some contemporary Roman brasses and are in no way inconsistent with these.

Cementation is the name of the process used in antiquity to manufacture brass and is quite different from the simple mixing of metals used to make bronze (Craddock 1995: 294-302). Because zinc metal boils at a lower temperature (907 °C) than that required for smelting (approx. 1100 °C) (unlike tin and most other metals), the process is essentially solid-state, where zinc vapour diffuses directly into copper metal. The process involved finely divided copper metal being packed into a closed crucible with charcoal and calcined zinc carbonate ore (smithsonite). The crucible was then placed in a furnace and heated to between 930 and 1000 °C. If the temperature were any lower the zinc ore would not reduce and produce vapour, any higher and the copper/brass would melt reducing the surface area and thus the reaction rate. The amount of zinc that the metal would absorb is essentially self-limited. Experimental work has also shown that brass produced by this process will contain a maximum zinc content of 28 % (Hedecke 1973). If scrap metal were being used in the cementation process, then the presence of any lead or tin in the copper would restrict the absorption of zinc. Consequently, early Roman brasses tend to be relatively ‘clean’ and free of significant tin and lead contamination. The relatively high zinc contents and lack of tin or lead in the Tel Anafa brass objects is therefore consistent with first century Roman brasses from both Israel (Ponting & Segal 1998) and from Europe (Craddock & Jackson 1995; Bayley 1998).

The lead contents of the copper alloys presents some interesting variations (Fig. 2). Lead is only present in significant amounts (above 2 or 3 %) in bronze objects, and this phenomenon is true for all three sites. Lead was commonly added to bronze alloys that were going to be used for casting because it lowers the viscosity and melting point of the alloy (Young 1967) and bronze tended to be the preferred alloy for casting. Brass, whether cast or hammered, generally contains negligible lead for the reasons discussed above. U-n alloyed copper was rarely used for casting in this period, often being used for hammered objects where the metal’s superior ductility was an advantage. U-n alloyed copper was also used for artefacts that were to be tinned or fire-gilded, where any alloying components would interfere with the plating process (Meeks 1993: 255; Oddy 1985). Furthermore, the addition of excessive amounts of lead to bronze has been identified as a characteristic of Late Hellenistic and Roman copies (Bayley 1985). Whilst lead is added to most bronze used for casting from the middle of the second millenium BC (Craddock 1985: 61), the amounts are seldom more than 10 % or so. Roman and Late Hellenistic cast bronzes consistently contain 20 % or more. It is therefore of interest to the theme of this paper that it is the bronzes from the pagan site of Tel Anafa that contain markedly higher levels of lead than either of the Jewish sites.

There are therefore two separate compositional features that mark the copper alloy objects from Tel Anafa out as different from the objects from Gamla and Yodfat. Most notably there are no brass objects from either Yodfat or the Gamla civilian assemblage. Secondly, the lead contents of the Tel Anafa
bronzes are also markedly higher than those found in the bronze objects from the other two sites. Both these characteristics relate to the major elements in the alloys, constituents that were manipulated by the artisans according to their requirements and traditions. The minor and trace elements, however, were not subject to conscious manipulation by the artisans because their presence was seldom noticed and only recognized where they had some impact on working properties. Even in such cases, the symptom caused by the presence of the impurity was rarely attributed to trace contaminants and was usually regarded as some special characteristic of the copper or bronze itself. Thus the minor and trace elements in an alloy can sometimes provide important information about the metals used, largely independent of conscious human manipulation, and potentially providing clues about the smelting and alloying technology as well as ore characterization.

In the case of the copper alloys from these three Galilean sites, the distributions of the minor and trace elements are fairly uniform. Surprisingly, the bronze and un-alloyed copper objects from Tel Anafa contain no significant traces of zinc, as may be expected if bronze and brass were being worked together. This, indeed, is the case for Late Pre-Roman Iron Age material in Britain (Dungworth 1996: 408) where zinc-contaminated bronzes become common, alongside proper cementation brasses. Thus we have to look for a different model, one where bronze and copper production is not in close enough proximity to brass production to allow contamination. One such model might be that the brass items were brought into Tel Anafa through trade contacts with the Romanised cities whereas bronze and copper continued to be worked on sites re-using scrap metal that had been circulating in the area for generations before any Roman contact. This model is certainly in agreement with the other Roman aspects of Tel Anafas' material culture such as the import of red-slipped table vessels and mould-made lamps (Berlin 2002). It would also seem likely that the import of brass objects was still a relatively new phenomenon tied into the increasing presence of the Roman army in the region. Consequently, it was not yet common enough for any brass objects to be recycled and therefore contaminate future copper alloy production. Alternatively, the high value and exotic appeal of the alloy may have led to different treatment and disposal of broken or damaged pieces.

The consistent distribution of the trace elements in all the copper alloy objects across the three sites lends some support to this discussion. When the trace element concentrations in the Galilean copper alloy objects are compared to those in Roman copper alloy artefacts there are some interesting differences. Figure 3 shows the arsenic and cobalt concentrations of the copper alloys from the three sites together with those measured in the Roman military copper alloy objects from Gamla and the contemporary siege site of Masada (Ponting & Segal 1998). All the data used here have been scaled to the copper in order to remove any dilution problems caused by the varying amounts of alloying components and to therefore make the data comparable. The plot shows that the Galilee copper alloys generally contain greater concentrations of both arsenic and cobalt, elements that are correlated and geochemically related to the types of copper ore originally smelted. Separation of the two groups is not total and there is considerable overlap of the Roman material that is probably explained by the recycling of local copper alloys by the Roman army. Nevertheless, the Galilee copper alloys have a reasonably tight distribution, a feature that is also demonstrated when the data are presented as box-and-whisker plots. Figure 4 shows the Box-and-Whisker plots for arsenic (a) and cobalt (b) and clearly shows the differences in the distribution of the trace elements in the Galilee and Roman copper alloys.

**Fig. 3.** Scatterplot showing the scaled arsenic and cobalt contents of the Galilee copper alloys compared to the Roman military copper-alloys.

**Fig. 4.** a) Box-and-Whisker plot of the scaled arsenic contents of the Galilee copper-alloys compared to the Roman military copper-alloys. b) Box-and-Whisker plot of the scaled cobalt contents of the Galilee copper-alloys compared to the Roman military copper-alloys. See caption to Fig. 2 for explanation.
made available by the excavators; Sharon Herbert and Gloria Merker (University of Missouri) for Tel A nafa, Motti Aviam (Israel Antiquities Authority) for Yodfat and Danny Syon (Israel Antiquities Authority) for Gamla. The Israel Antiquities Authority kindly granted permission for the sampling and special thanks are due to Pnina Shor for facilitating access to material. David Taylor (University of Nottingham) kindly drew Figure 1.

References
Ponting, M. J. forthcoming c. ‘Chemical analysis of copper-alloy metalwork from the Roman context’. In: S. Her bert (ed), Tel A nafa Vol. 3. Journal of Roman Archaeology Supplementary Series.
Ponting, M. J. forthcoming d. ‘Chemical Analysis of the Copper Alloys from the Early Roman Period’. In: M. A.viam (ed), Excavations at Yodfat, University of Rochester.

Acknowledgments
This project was initiated through many e-mail discussions with Andrea Berlin (University of Minnesota) and Danny Syon (Israel Antiquities Authority). Material was generously

Conclusion
Berlin’s identification and study of the features of Galilean material culture used to signal cultural identity and affiliation have provided the starting point for a more technologically oriented study. The use of brass by the pagan population of the Galilee was in line with other groups on the periphery of and in contact with Roman culture, as demonstrated in Britain (Bayley 1998) and Gaul (Hamilton 1996). The fact that the Jewish population deliberately chose not to ‘buy into’ this, and treated objects of brass with the same disdain that they showed to red-slipped pottery and mould-made lamps, suggests that brass was viewed as a distinctly Roman product.

Social scientists have shown that certain groups who live in areas of culturally mixed populations will deliberately adopt ‘identity-signaling’ features. Such features are almost always material rather than behavioral (Stevenson 1989: Moore’ 1987). There appears to be no set rule to this; people chose to reject or adopt those features around them that they feel advertise their cultural affiliation. A S Berlin puts it, “people will make a statement with whatever they can, when they feel the need to do so.” (Berlin 2002).

This observation reinforces the assessments made by Bayley (1998) that the manufacture, working and use of brass is, in some quite fundamental way, linked to Rome in the minds of the members of populations peripheral to the Roman provinces. Furthermore, the presence of significantly higher levels of lead in the Tel A nafa bronzes than in the bronzes from the Jewish sites suggests a difference in approach to casting technology. The use of highly leaded bronze is consistent with most Late Hellenistic and Roman cast metalwork (Craddock 1985; Beck et al. 1985), whereas the use of, at most, the smallest amount of lead necessary to improve the alloy’s casting properties is a feature of earlier Near Eastern metalurgy (M oorey 1994: 263). This, again, points to significant differences in the attitudes of the two populations, one that suggests differences in approaches to technology as well as material culture.

Interestingly, the trace element distributions indicate that both groups were drawing on a generally homogeneous copper-alloy supply pool. In fact two of the brass objects from Tel A nafa have levels of arsenic (when scaled) consistent with the local Near Eastern copper and therefore suggests that local copper was being used to produce brass, a suggestion that has been aired elsewhere (Ponting forthcoming a). The small size of the sample obviously necessitates caution in interpreting these results and more data will be needed before such statements can be substantiated.

Furthermore, this project demonstrates that the study of the selection, modification or exclusion of every type of material used to manufacture material culture is an important tool in the study of past societies. These elements between the Galilee and Roman metalwork. Furthermore, the greater tendency for the Roman distributions to include relatively large numbers of outliers and extreme values is also demonstrated, a feature responsible for the overlap apparent in the scatterplot. Significantly, the brass objects from Tel A nafa all have low levels of cobalt, yet the arsenic concentrations divide the four objects into two distinct groups. Both of the fibulae contain negligible amounts of both cobalt and arsenic, whereas the cosmetic implement and the bracelet contain significant (>0.2 %) levels of arsenic. The suggestion must be that the fibulae are likely to be Roman military products, whereas the other items may represent Near Eastern brass production. It is therefore worthwhile noting that both fibulae are auscissa types, a form known to be particularly favored by the Roman army (F eugère 1985).


Matthew Ponting, Keeping up with the Romans?
The Cave of the Sandal, Ketef Jericho: new evidence from recent Chalcolithic copper finds

Irina Segal, Andrei Kamenski & John Merkel

Introduction

In 1993 during ‘Operation Scroll’, two copper tools and one copper mace-head were found at the Cave of the Sandal, Ketef Jericho (Eshel & Zissu 1998; 2000). The ‘utilitarian’ tools represent two different Chalcolithic types: a flat axe and a narrow chisel. The disk-shaped, undecorated mace-head is also a known Chalcolithic metal artefact type, but the disk-shape is not the most common variation (Bar-Aron 1980). Mace-heads probably served foremost as weapons. However, it is possible that mace-heads also encompassed some additional symbolic meanings, as a prestige object, at contemporary sites throughout the wider region. Sufficiently large numbers of the copper and/or copper-alloy artefacts from the Chalcolithic period have been discovered in excavations in the Southern Levant (Ilan & Sebbane 1989; Levy & Shalev 1989; Gonen 1991; Levi 1998) to build a comprehensive typology of metal objects. The technical investigations of these three artefacts from the Cave of the Sandal, requested by the excavators Dr. Hanan Eshel and Boaz Zissu, provide new compositional and metallurgical data against which to compare metal object types from other Chalcolithic sites.

Alloy compositions cannot be evaluated accurately without undertaking quantitative compositional analysis of uncorroded metal remaining in the artefacts, but not all objects are available for sampling. Selected Chalcolithic copper artefacts have already been investigated for composition and metallographic structures, such as the ‘treasure’ of Nahal Mishmar (Key 1980; Potashkin & Bar-Avi 1980; Shalev & Northover 1991; Tadmor et al. 1995) as well as metal artefacts from Nahal Zeelim (Key 1980; Shalev & Northover 1993), Bir Safadi (Tylecote, Rothenberg & Lupu 1974), Shiglim (Shalev & Northover 1987), Nahal Makuh, Nahal Qanah and Gilat (Shalev & Northover 1995) and Peqi’in cave (Segal et al. in prep.). Based upon published compositional data, it has been proposed that during the Chalcolithic period two distinct metal ‘industries’ existed to produce copper tools and prestige objects (Levi 1998). Unexpectedly, the utilitarian tools were made of unalloyed copper while the prestige objects, such as crowns and standards, were made of copper alloyed with arsenic and/or antimony. Normally, one would expect tools to be alloyed in order to produce more effective cutting edges, taking advantage of the higher hardness produced by cold-working/copper-arsenic alloys, for example. Nevertheless, this is not the case. Rather than hardness, then, other properties, perhaps colour and better casting characteristics, of copper-arsenic or copper-antimony alloys were preferred, and possibly deliberately selected for specific prestige metal objects.

The aim of the current research was to investigate the metal composition, metallography and microhardness of the three excavated artefacts. Are the axe and chisel unalloyed? Is the mace-head also unalloyed? If so, then the mace-head should be classified, on composition alone, perhaps more as a functional tool than a prestige object. This technical investigation aims to reconsider briefly the classification of Chalcolithic tools and weapons, and contrast the results with published technical data for prestige metal objects. The production of copper-arsenic alloy objects has been viewed as most probably quite distant, outside the Southern Levant (Levy 1998). Nevertheless, several other options exist for procedures and locations for production of copper-arsenic alloys (Tylecote 1991). In addition, we tried to assess closer locations of ores possibly utilised for artefact production. Lead isotope ratio determination is very useful for provenance studies, and was used here for this end.

Analytical procedures

Samples for metallography were cut with a jeweler’s saw. The sections were mounted in epoxy resin, ground and polished following usual procedures. The polished sections were etched in aqueous ferric chloride and acidified potassium dichromate solutions. The metallographic structures were observed under a Nikon metallurgical microscope.

The mounted sections were also subjected to Vickers micro-hardness tests to quantify better the combined effects of cold-working/annealing and alloy compositions. The load used was 300 g. Microhardness values and the estimated degree of cold-working are especially important for evaluating tools of unalloyed copper as well as prestige objects of selected copper alloys.

The metal compositions of the three artefacts were determined by Inductively Coupled Plasma Atomic Emission Spectrometry (ICP-AES) using Jobine Y von JY -38 mono- and JY -48 poly-chromators at the Geological Survey of Israel. The operating conditions of the ICP-AES have been described earlier (Segal et al. 1994). The limits of detection (LOD) of determined elements are estimated at about 1 ppm for the selected sample size and dilution of the sample solution. In addition, the artefacts were drilled, with the initial corrosion products excluded from the 25 mg samples. The precision, or relative standard deviation (RSD) of method for the analyses is as following: 1 % for major elements, about 3 % for minor elements and about 10 % for trace elements. The results of the ICP-AES were checked against area analyses under Scanning Electron Microscope (J EOL 840) equipped with an Energy Dispersive System (SEM - EDS) for the metallographic sections. A good accordance was observed between the two analytical techniques. Detailed micro-analytical and metallographic studies were performed on the mounted samples using SEM - EDS and a back-scattered electron detector (BE1). A mount of oxygen inclusions was estimated under a Nikon metallurgical microscope using polished sections.

Lead isotope determination was carried out using a Perkin-Elmer Sciex Elan 6000 Inductively Coupled Plasma Mass Spectrometer (ICP-MS). The precision of this method for major lead isotopes is about 0.3 % (Halicz, Erel & Veron 1996).

Copper artefacts

Drawings of the three artefacts as well as their sizes and selected sample positions are shown in Fig. 1. The Locus (L)
and Basket (B) numbers designate the excavation location and object find number in the publication of the site. The objects were relatively well-preserved, only copper oxide and carbonate corrosion layers covered their surface. From the metallographic sections, the penetration of corrosion into the metal is estimated at about 30 microns.

1. Flat axe (L 30, B 165) with convex surfaces and sharp rounded edge. Its weight is 285.5 g, its length is 13.0 cm. A mounted metallographic section was made from the cutting edge (transverse section). No analogs to this axe were found in Bar-Adon (1980).

2. Narrow chisel (L 30, B 171) with widening towards its sharp rounded working edge. It was broken in antiquity, but both parts were found in the excavations. Its weight is 229.5 g, its length 12.1 cm. The chisel was sampled from the cutting edge both in transverse and longitudinal sections. Fifteen chisels found in Nahal M ishmar varied in length from 15 to 30 cm and in weight from 156 to 807 g (Bar-Adon 1980).

3. Disk-shaped mace-head (L 30, B 1101) with convex surfaces and shaft-hole, the rims of which have sharp carinated edges. It weighs 242.5 g and is 8.5 cm in diameter. The mace-head was sampled twice: from the disk edge and from the carinated end of the shaft-hole in transversal sections. Bar-Adon (1980) called this object a ‘disk’ and grouped it together with short standards. Eleven ‘disks’ from Nahal M ishmar ranged in weight from 103 to 282 g and in diameter from 5.6 to 7.0 cm (Bar-Adon 1980).

Analytical results
Overall compositional analysis by ICP-AES (Table 1) shows that each artefact is made of unalloyed copper. The two tools are significantly "pur"er than the mace-head, they contain less than 0.1% of impurities. Only traces of iron, cobalt, manganese and lead were observed in the tools. The disk-shaped mace-head contains higher concentrations of lead (0.013 %), nickel (0.17 %) and iron (0.015 %). These observations are in correspondence with published results on Nahal M ishmar objects. It is confirmed again that (so far) all the Chalcolithic tool-types were made of unalloyed copper (Shalev & Northover 1993; 1995; Tadmor et al. 1995). In contrast, according to Key (1980) and Tadmor et al. (1995), although most of prestige objects were of alloyed copper, two standards (61-52 and 61-104) were also found to have been made of unalloyed copper. It is possible that Nahal M ishmar artefacts 61-40, 61-58 and 61-426 with arsenic content less than 1.92 % (Key 1980) are also unalloyed, because this value corresponds to the limit of detection of the optical-emission analytical technique used by Key. Now, the disk-shaped mace-head from the Cave of the Sandal is also shown to be unalloyed copper and probably should not be classified together with ‘short standards’ as done by Bar-Adon (1980). Therefore, a proposed distinction between unalloyed tool-types and alloyed prestige objects is not necessarily exclusive. More objects should be investigated to assess the significance of this proposed correlation. Cold-working properties and marginal colour differences are readily apparent for production of sheet metal of copper-arsenic alloys (Lechtman 1996). However, intermediate alloy colours and compositions around 1-2 % arsenic in copper may not be very distinctive for larger, cast, partially cold-worked, utilitarian tools. Prestige copper-arsenic alloys with over 2 % arsenic are strikingly different in colour. Furthermore, loss of arsenic is most probable from remelting under oxidising conditions (McKerrell & Tylecote 1972). Thus, some overlap and mixing of alloy compositions and object types should actually be expected. Based upon composition alone, the three artefacts analysed from the Cave of the Sandal could have been smelted from high-grade copper ores; such as the best examples of copper ore from Feinan as well as the Timna region. These variable copper ores can also contain significant concentrations of iron and sulphur. Sometimes nickel is detected in ores. It has been proposed that up to 0.4 % of nickel in Abu Matar and Bir es Safadi axes originated in Feinan ores (Hauptmann 1989) and 0.15 % of nickel in the Timna copper (Rothenberg 1990).

Table 1. Chemical composition of copper artefacts from the Cave of the Sandal, in wt %.

<table>
<thead>
<tr>
<th>Identification</th>
<th>Sn</th>
<th>Zn</th>
<th>As</th>
<th>Sb</th>
<th>Pb</th>
<th>Co</th>
<th>Ni</th>
<th>Fe</th>
<th>Mn</th>
<th>Ag</th>
<th>Au</th>
<th>Cu</th>
</tr>
</thead>
<tbody>
<tr>
<td>Axe</td>
<td>nd</td>
<td>nd</td>
<td>nd</td>
<td>nd</td>
<td>nd</td>
<td>nd</td>
<td>0.035</td>
<td>0.005</td>
<td>0.0001</td>
<td>0.015</td>
<td>nd</td>
<td>99.2</td>
</tr>
<tr>
<td>Chisel</td>
<td>nd</td>
<td>nd</td>
<td>nd</td>
<td>nd</td>
<td>0.009</td>
<td>0.001</td>
<td>0.045</td>
<td>0.006</td>
<td>0.0003</td>
<td>0.006</td>
<td>nd</td>
<td>98.9</td>
</tr>
<tr>
<td>Disk-shaped mace-head</td>
<td>nd</td>
<td>nd</td>
<td>nd</td>
<td>nd</td>
<td>0.013</td>
<td>0.001</td>
<td>0.17</td>
<td>0.015</td>
<td>0.0003</td>
<td>0.015</td>
<td>0.001</td>
<td>99.1</td>
</tr>
</tbody>
</table>

Note: nd - not detected.

Metallography and microhardness measurements
Unalloyed, annealed copper has a Vickers microhardness of about 40 H V, while cold-worked copper at about 95 % reduction in thickness reaches a microhardness maximum at around 120 H V as kg/mm² (Smith 1982: 95). In practical terms, these differences are readily observed for copper sheet and for edge hardness. One might expect copper ‘utilitarian’ tools to have appreciably hardened cutting edges.

1. Flat axe
Visual examination of the axe revealed ‘stipples’ on both sides of the working edge, suggesting its use for hard materials. The
polished section revealed tiny copper sulfide inclusions located at the boundaries of the initial casting grains. They range in size from 0.09 to 0.15 mm. The copper contains less than 0.03 % of oxygen. Clear structure appears after etching (Fig. 2). The etched sample showed a similar structure in both sections: slightly deformed recrystallised polygonal grains (size 0.02-0.04 mm) superimposed on compressed original grains (Fig. 5). The compression degree increases towards the blade from 20-40 % up to 50-60 %. The structure is not fully recrystallised.

The axe was cast in a two-piece closed mould. After casting, it was cold-worked and annealed at 700-800 °C, forging was finished on cooled metal. Microhardness was measured at 100 HV and 108 HV for the blade edge. A gain, the working edge was work-hardened. This narrow chisel could serve as a suitably utilitarian tool.

3. Disk-shaped mace-head
Visual observation of the surface defects indicated that a two-piece closed mould was used for casting. This mace-head does not contain a cavity inside; it is a solid casting. Both polished sections revealed the presence of copper sulfide and Cu-Cu2O inclusions around the casting grains. The sample from the shaft-hole edge contains less than 0.01 % of oxygen and the second one 0.05-0.07 %. In both the etched sections partly recrystallised, undeformed, small grains superimposed on deformed elongated cast grains were observed (Fig. 6). Their compression is estimated at about 40 %.

The disk was cast and subsequently hot-forged at ca. 400 °C to remove casting defects. The shaft-hole edge was slightly cold-worked. The microhardness of the disk edge was 95 HV and near the shaft, 98 HV. These microhardness values most likely represent surface ‘finishing’ of the disk possibly as well as deliberate work-hardening of the edge and around the shaft-hole. Due to the limited section size and depth into the object, the center microhardness could not be measured for better comparison.

Provenance study
For provenance elucidation of the three studied objects, lead isotope ratios were measured. 207Pb/206Pb and 208Pb/206Pb ratios are shown in Table 2 and Fig. 7. For comparison of our data with the relevant ores, results of lead isotope ratios in Feinan (Hauptmann et al. 1992) and Timna (Gale et al. 1990) ores are also plotted. Values for Feinan ores are indistinguishable from Timna ores. A was concluded by Hauptmann et al. (1992) two types of copper ores, Middle Brown Sandstones (M BS) and Dolomite Limestone Shales (DLS), were used for ancient smelting at Feinan. M BS plot in the lower left area of the isotope graph, values of DLS are significantly higher. The isotope ratios of Chalcolithic objects from Feinan fall into a wide range showing that both types of ore were used in the Chalcolithic period. Later - from EB II - only DLS were utilised. Timna copper ores, having mainly higher lead isotope ratios (Gale et al. 1990), cover only the Feinan DLS range. A comparison of lead isotope ratios for our objects with published Chalcolithic and early stage EB I copper artefacts from Levant is shown in Fig. 8. The data used

<table>
<thead>
<tr>
<th>Identification</th>
<th>207Pb/206Pb</th>
<th>208Pb/206Pb</th>
</tr>
</thead>
<tbody>
<tr>
<td>Axe</td>
<td>0.8424</td>
<td>2.0819</td>
</tr>
<tr>
<td>Chisel</td>
<td>0.8383</td>
<td>2.0815</td>
</tr>
<tr>
<td>Disk-shaped mace-head</td>
<td>0.8613</td>
<td>2.1059</td>
</tr>
</tbody>
</table>
to emphasise the existence of local metalworking and object typology as well as alloy selection in the Chalcolithic period.

Acknowledgements
We would like to thank the excavators, Dr. Hanan Eshel and Boaz Zissu for the opportunity to analyse these interesting artefacts. The structures of the metallographic samples were interpreted on the basis of copper standard samples prepared, hot-worked and investigated in the laboratory of Prof. N. Rynida at Moscow University.

References
Chemical composition of prehistoric copper artefacts from Transylvania, Romania

Manuela Kadar

Abstract
This work is part of a large archaeometallurgical project, which aims to study the beginnings of metalworking in Transylvania. Seventeen samples of Eneolithic and Bronze Age objects were analysed by Inductively Coupled Plasma - Atomic Emission Spectrometry and Inductively Coupled Plasma - Mass Spectrometry. The analyses disclosed high-purity copper for most of the heavy artefacts, a fact which has opened the discussion on distinguishing between native copper and smelted copper as raw material used for manufacturing Eneolithic artefacts.

Introduction
Transylvania is referred to in the literature as an important region for the prehistory of central and south-eastern Europe, being well known for its rich deposits of gold, copper and salt, which were exploited from early times. The beginnings of metal production in this area still raise a lot of questions as concerns the nature of raw materials, extractive activities, processing technologies, circulation of heavy artefacts, and intentional alloying of copper with arsenic and antimony. With regard to the development of independent metallurgical centres in the south-east of Europe, Chernykh (1978; 1992) defined a Balkano-Carpathian metallurgical province with several subprovinces and related them to the production centres of Ai Bunar, Rudna Glava and an unknown centre in the Carpathian Mountains. This concept is based on the optical emission spectroscopy analyses of typologically different heavy Eneolithic artefacts. According to Chernykh, differences in typology are accompanied by differences in raw material composition, such as the concentrations of trace elements like arsenic, nickel, silver and gold. Todorova has completed the previous work with a detailed typological study and defined three metallurgical sub-provinces in south-eastern Europe, namely Transylvania, Serbia and parts of Dalmatia and Thrace (Todorova 1981; Pernicka et al. 1997).

While the Serbian and Bulgarian metallurgical centres have been described in monographic studies, in Transylvania such a systematic, multidisciplinary project for the identification of prehistoric mines and complex analytical studies of the Eneolithic and Early Bronze Age artefacts was never carried out. The prehistoric copper mines from Rudna Glava, Bor and Mădăianpek in Serbia and Ai Bunar in Bulgaria and their relationship to the artefacts discovered in the nearby area have been investigated by interdisciplinary projects. There have been established comparisons between the chemical composition and/or isotopic composition of the sources of raw materials and archaeological finds (Pernicka et al. 1993; Begemann et al. 1995; Pernicka et al. 1997; Jovanovic 1976; Jovanovic 1978; Todorova 1999). This approach has in many cases rejected the simplistic standard geographical proximity of the raw sources considered in the existing archaeological models.

Transylvania has yielded an impressive amount of heavy copper objects (altogether 500 kg), objects belonging to Eneolithic cultures such as Petreseşti, Tiszapogá, Bodrogkereszter, Herculeane-Čeile and Türzii-H unyadhalom. These artefacts have probably been produced by the exploitation of copper ore deposits from the Carpathians (Vulpe 1973: 220-234; Comşa 1987: 102-109). Evidence of local extraction and primary metallurgy of copper have motivated us to start an archaeometallurgical investigation within the Carpathian Arch. This project aims to establish the origin of raw materials used for the manufacture of Eneolithic and Early Bronze Age copper artefacts and to study the applied technologies within the Eneolithic and Early Bronze Age cultures of Transylvania, in comparison with technologies used in other regions from south-eastern and central Europe. We have started in 1997 with the creation of a relational database system of copper ores from the Carpathians (Kadar 1999). The database has been continuously updated with Eneolithic and Early Bronze Age archaeological finds from Transylvania. A valuable analytical data has been entered into the database and further investigations have been carried out as concerns the chemical composition and metallographic structure of the objects in discussion (Kadar 2000).

This paper presents new data on the chemical composition of the heavy implements, mainly from south-western Transylvania. Analyses were carried out by Inductively Coupled Plasma - Atomic Emission Spectroscopy (ICP-AES) and Inductively Coupled Plasma - Mass Spectrometry (ICP-MS) at the European Large Scale Geochemical Facility, University of Bristol, Great Britain.

Catalogue of archaeological materials studied
The artefacts studied here belong to the south-west of Transylvania, a region very rich in Eneolithic metal finds (Figure 1).

Methodology
Seventeen samples were analysed by both ICP-AES and ICP-MS. Bulk chemical analyses were carried out by ICP-AES and trace elements such as Ag, Co, Ni, Hg, Zn, Au, Pb were measured by ICP-MS. The advantages of both methods are an overall good precision, accuracy and rapidity when compared to other analytical techniques such as neutron activation or atomic absorption spectrometry.

Samples were removed from each artefact by drilling a one millimetre diameter hole and collecting the turnings. The first two or three millimetres of material were discarded to exclude metal corroded by inter-granular corrosion or chemically altered by corrosion processes. Only one hole...
was permitted in each artefact thus the place of sampling had to be carefully chosen in order to be representative for the object. Where the object to be sampled was too thin for drilling, a scalpel was used to scrape a sample from an edge, which had been previously cleaned mechanically.

Sample preparation started with cleaning in excess of acetone, then 50 mg of each sample was weighed accurately into a PTEF beaker, 5 ml of aqua regia (HCl+HNO₃ mixture, 3:1) were added and then made up to 25 ml volume using deionised water. Some of the samples were microwave digested in order to keep possibly volatile trace elements in the solution. Determinations were made using an Jobin Yvon JY 24 instrument for supposed major elements such as Cu, As, Fe, Sb and trace elements were determined by ICP-MS on a PlasmaQuad equipment. Synthetic multi-element standards were made up from commercial standard solutions. A series of five standards and a blank were made up to run first and to collect the count data for each concentration. Then the samples were run with standard reference materials to monitor the accuracy of the measurements and these were prepared in exactly the same way as the artefacts.

The precision of data (the relative standard deviation) is estimated at ± 1 % for major elements and for trace elements around ± 10 %. The wavelength used to analyse copper was 324.754 nm.

Results and discussions
One of the most debated problems is the nature of raw material used for the production of the Transylvanian Eneolithic and Early Bronze Age artefacts. To understand the early use of copper we have tried to identify objects as having been made from native copper and to distinguish them from objects made from smelted copper. Two techniques have been used in combination: trace element analyses and microstructure characterisation.

Here we will discuss only the chemical composition of the samples presented in Table 1. Some objects have been already analysed by emission spectroscopy at the Württembergisches Landesmuseum of Stuttgart (Junghans et al. 1974) and by neutron activation (Beşliu et al. 1992; Beşliu & Lazarović 1995). As regards the Eneolithic artefacts, two of them, B1 and B12 present traces of mercury in the composition. The presence of mercury (0.62 ppm and 2.16 ppm), together with low concentrations of cobalt and nickel as impurities, would suggest that these objects have been made by cold hammering and possibly tempering of native copper. As stated recently by Pernicka et al. (1997: 118): "Smelting of copper removes mercury very effectively so this element might possibly serve as a useful indicator to distinguish between artefacts made of native copper and such made from smelted copper".

Other useful markers to distinguish between native and smelted copper are cobalt and nickel which are almost invariably higher in smelted copper, as indicated by analyses of copper prills found in Chalcolithic and Early Bronze Age slags from Feinan, Jordan (Hauptmann et al. 1992).

With regard to arsenic, one object (B4) presents higher concentration (7114 ppm), otherwise arsenic contents are much lower. We conclude that arsenic was not added deliberately but was a natural impurity in the ores used. The same is true
Looking at iron, according to Junghans et al. (1974), an iron content below 1% seems to be typical of Eneolithic and Early Bronze Age artefacts. The low content of iron may have several reasons. It may be caused by smelting rich oxidic copper ores, i.e., relatively pure malachite with a low content of gangue from the matrix rock, or an originally high content of iron may have been reduced by a technological process. Another reason would be the remelting before casting, with the oxidation and slagging of iron as demonstrated experimentally by Tylecote and Boydell (1978) and by Merkel (1990).

The problem of distinguishing between native and smelted copper when dealing with high purity copper from south-eastern Europe has been raised in the last century, further studied by Otto and Witter (1952) by optical emission spectroscopy and presented by Junghans and co-workers in their study of prehistoric metal objects from all over Europe. These scholars are inclined to state that spectroscopically high purity copper in objects is indicative of native copper as source. Chernykh (1992) states that his chemical groups I and II derive from monometallic copper ores.

In Transylvania, evidence concerning the early smelting of copper ores starts with Eneolithic cultures and continues in the “transition period” and in the Early Bronze Age cultures. For example, one of the finds, a piece of melted metal discovered at Livezile-Alba County, in the context of the chronological horizon of the Livezile group of the Early Bronze Age, has the chemical composition of an alloyed copper: 91.8% Cu, 7.2% Zn, 0.8% Fe, 0.1% Pb and 0.03% As. Local communities have overcome the phase of processing native copper and have started to exploit local complex ores in which copper is associated with zinc, such as ores from Bucium and Sârâm (Ciugudean 1996: 119; Beşliu et al. 1992: 123, appendix 4).

For antimony when talking about the Eneolithic objects.

Table 1. Chemical composition of copper artefacts from Transylvania, Romania, obtained by ICP-AES and ICP-MS analyses.

<table>
<thead>
<tr>
<th>Sample ID</th>
<th>Description</th>
<th>Period</th>
<th>Location</th>
<th>Cu wt%</th>
<th>As ppm</th>
<th>Fe ppm</th>
<th>Pb ppm</th>
<th>Sn ppm</th>
<th>Ag ppm</th>
<th>Co ppm</th>
<th>Ni ppm</th>
<th>Hg ppm</th>
</tr>
</thead>
<tbody>
<tr>
<td>B1</td>
<td>Small Chisel</td>
<td>Eneolithic</td>
<td>Şteaua-Gorgan Alba</td>
<td>96.2</td>
<td>79</td>
<td>284</td>
<td>11</td>
<td>11</td>
<td>90</td>
<td>41</td>
<td>4</td>
<td>75</td>
</tr>
<tr>
<td>B2</td>
<td>Axe-adze</td>
<td>Eneolithic</td>
<td>Sebeş-Alba</td>
<td>97.6</td>
<td>89</td>
<td>52</td>
<td>68</td>
<td>9</td>
<td>71</td>
<td>32</td>
<td>&lt;2</td>
<td>75</td>
</tr>
<tr>
<td>B3</td>
<td>Axe-adze</td>
<td>Eneolithic</td>
<td>Petreşti-Alba</td>
<td>100.6</td>
<td>84</td>
<td>38</td>
<td>24</td>
<td>11</td>
<td>77</td>
<td>92</td>
<td>&lt;2</td>
<td>42</td>
</tr>
<tr>
<td>B4</td>
<td>Shaft-hole axe</td>
<td>Bronze Age</td>
<td>Șișa-Salaj</td>
<td>95.3</td>
<td>71</td>
<td>14</td>
<td>62</td>
<td>247</td>
<td>869</td>
<td>123</td>
<td>720</td>
<td>2</td>
</tr>
<tr>
<td>B5</td>
<td>Hammer axe</td>
<td>Eneolithic</td>
<td>Turda-Alba</td>
<td>93.6</td>
<td>41</td>
<td>24</td>
<td>51</td>
<td>4</td>
<td>25</td>
<td>75</td>
<td>4</td>
<td>8</td>
</tr>
<tr>
<td>B6</td>
<td>Axe fragment</td>
<td>Eneolithic</td>
<td>Ghirbom-Alba</td>
<td>96.6</td>
<td>125</td>
<td>ND</td>
<td>22</td>
<td>26</td>
<td>69</td>
<td>136</td>
<td>18</td>
<td>ND</td>
</tr>
<tr>
<td>B7</td>
<td>Axe-adze</td>
<td>Eneolithic</td>
<td>Alba</td>
<td>98.4</td>
<td>25</td>
<td>6</td>
<td>9</td>
<td>3</td>
<td>76</td>
<td>50</td>
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<td>79</td>
</tr>
<tr>
<td>B8</td>
<td>Axe-adze</td>
<td>Eneolithic</td>
<td>Șoâlăni-Alba</td>
<td>97.7</td>
<td>22</td>
<td>14</td>
<td>16</td>
<td>9</td>
<td>8</td>
<td>70</td>
<td>59</td>
<td>&lt;1</td>
</tr>
<tr>
<td>B9</td>
<td>Knife blade</td>
<td>Eneolithic</td>
<td>Sibiu-Alba</td>
<td>97.1</td>
<td>35</td>
<td>27</td>
<td>39</td>
<td>16</td>
<td>165</td>
<td>49</td>
<td>79</td>
<td>56</td>
</tr>
<tr>
<td>B10</td>
<td>Axe-adze</td>
<td>Eneolithic</td>
<td>Cetatea de Baţa-Alba</td>
<td>97.9</td>
<td>17</td>
<td>16</td>
<td>454</td>
<td>449</td>
<td>&lt;3</td>
<td>11</td>
<td>79</td>
<td>33</td>
</tr>
<tr>
<td>B11</td>
<td>Axe-adze</td>
<td>Eneolithic</td>
<td>Ormeni-Alba</td>
<td>97.3</td>
<td>6</td>
<td>34</td>
<td>449</td>
<td>421</td>
<td>&lt;3</td>
<td>5</td>
<td>67</td>
<td>30</td>
</tr>
<tr>
<td>B12</td>
<td>Axe fragment</td>
<td>Eneolithic</td>
<td>Cetatea-Alba</td>
<td>98.4</td>
<td>15</td>
<td>548</td>
<td>437</td>
<td>453</td>
<td>&lt;3</td>
<td>34</td>
<td>77</td>
<td>157</td>
</tr>
<tr>
<td>B13</td>
<td>Chisel</td>
<td>Bronze Age</td>
<td>Rămeşti-Alba</td>
<td>93.0</td>
<td>257</td>
<td>2840</td>
<td>1646</td>
<td>1286</td>
<td>99300</td>
<td>24</td>
<td>295</td>
<td>1736</td>
</tr>
<tr>
<td>B14</td>
<td>Copper ingot</td>
<td>Hallstadt</td>
<td>Șoâlăni-Alba</td>
<td>95.7</td>
<td>108</td>
<td>2010</td>
<td>585</td>
<td>568</td>
<td>&lt;2</td>
<td>17</td>
<td>146</td>
<td>33</td>
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<tr>
<td>B15</td>
<td>Axe fragment</td>
<td>Eneolithic</td>
<td>Ungureni-Alba</td>
<td>102.3</td>
<td>6</td>
<td>10</td>
<td>417</td>
<td>397</td>
<td>&lt;2</td>
<td>5</td>
<td>93</td>
<td>55</td>
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<tr>
<td>B16</td>
<td>Copper ingot</td>
<td>Late Bronze Age</td>
<td>Cetatea-Alba</td>
<td>91.7</td>
<td>19420</td>
<td>43</td>
<td>3577</td>
<td>22417</td>
<td>22460</td>
<td>39</td>
<td>211</td>
<td>3691</td>
</tr>
<tr>
<td>B17</td>
<td>Knife blade</td>
<td>Bronze Age</td>
<td>Capuţi-Alba</td>
<td>98.3</td>
<td>&lt;2</td>
<td>27</td>
<td>471</td>
<td>488</td>
<td>&lt;3</td>
<td>19</td>
<td>205</td>
<td>107</td>
</tr>
</tbody>
</table>

ND = not detected.
nickel between 18 and 75 ppm and concentrations of cobalt of less than 8 ppm, with one exception for cobalt (sample B12 in which the concentration is 49 ppm).

Conclusions
Our research will be continued and extended to the eastern part of Transylvania in order to identify possible geological sources of raw materials in the Eneolithic and EBA and to reconstruct production and exchange systems at the level of Eneolithic and EBA communities. By updating our archaeometallurgical database with new analyses and finds we would like to fill, at least in part, a gap in the documentation of Romanian prehistoric metallurgy.

Acknowledgements
We acknowledge the support of the European Community Human Potential Programme, contract HPR-CT-1999-00008 awarded to Prof. B.J. Wood (EU Geochemical Facility, University of Bristol). The archaeological finds were made available by the History Museum of Alba Iulia, Dr. Horia Clugudean, the History Museum of Transylvania Cluj Napoca, Dr. Gheorghe Lazarovici, the History Museum of Sebeş, M. r. Marcel Simina, and the History Museum of Aiud, M. r. Paul Scrobota.

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Three new Aguada bronze plaques from Northwest Argentina

E.D. Cabanillas, L.R. González and T.A. Palacios

Abstract

In the Andean region of Northwest Argentina, the Prehispanic societies developed a unique metallurgical tradition. During the Period of Integration, 500 AD - 900 AD, artisans belonging to the well-known Aguada society, produced different types of bronze ornaments. Because of their rarity and complex manufacture, the decorated plaques transcend representations of high symbolic value. Few technical laboratory analyses made on this kind of objects are known. In this work we report laboratory studies done on three specimens of this type of plaques, and discuss their manufacturing process.

Metallurgy in prehispanic Northwest Argentina

The Indian societies that inhabited Northwest Argentina developed, from the 10th century BC until the arrival of the Spanish in the early 16th century, a sophisticated metallurgical production, which gave the region a particular identity in the Andean area (see Fig. 1). A small quantity of gold and silver objects is known, but the majority are made of copper or copper alloy, even with zinc (A.R. González 1979, L.R. González 1999). The archaeological data indicates that the smelting of ores and the preparation of alloys were firmly established by the 3rd century AD in the central east of the Catamarca province. The technology would have initiated and developed in total independence from the known metallurgical centres of South America, such as the Peruvian Andes and the Bolivian plateau (L.R. González 1994; A.R. González 1998: 94-95; cf. West 1994: 7). In the following centuries the metal production increased both in number of objects and in the volume of metal involved per object. In the time immediately preceding the European invasion, the artisans made tin-bronze ornamental pieces of up to two kilogram each, like disks and oval-formed section bells.

In this development an inflection point seems to have happened during the Integration Period (500-900 AD), hegemonized by the La Aguada socio-cultural entity. A n assortment of villager communities shared a complex religious system, which included ceremonial centres with pyramidal structures and blocks with human sacrifices and consumption of hallucinogens. Aguada developed itself in the central east of the Catamarca, but had a much wider influence over a large space of Northwest Argentina. Besides, sumptuary items moved across different boundaries as far as the Chilean North (Pérez Gollán 2000).

The increase in power of the political and religious leaders and the differentiation of their status at the interior of the society led to the formation of artisan groups specialised in the production of goods of excellent quality. One of the best known achievements of the artisans from Aguada is the pottery with religious scenes decorated with the anthropomorphous “The sacrificer”, felines and saurians. But artisans demonstrated a unique mastery of metallurgical work too.

For some time we have been developing a research program about the Northwest Argentinean prehispanic metallurgy (Cabanillas et al. 1998; L.R. González & Palacios 1996; Ziobrowski et al. 1996; Palacios et al. 1998; L.R. González et al. 1998, 1999a and b) connected with the explanation of technological characteristics of different metallic materials. The obtained data increased the knowledge not only about the trajectory of the metallurgical activity in the region but also the dialectics between the technology and the social processes (Oramam 1998). Related to this, it can be affirmed that in prehispanic Northwest Argentinean metallurgy, production was principally directed at ornaments, religious and ostentation of prestige positions in the communities (L.R. González...
& Peláez 1999). This situation was particularly noticeable in the context of the Aguada society (A.R. González 1998: 367).

**The Plaques in the Integration Period**

The Aguada plaques stand out because of their relative scarcity (only about thirty pieces are known), their detailed decoration and the symbolic charge in their iconography. The majority of these objects do not come from archaeological excavations and this is a problem for scientific studies. Furthermore, many of the pieces are isolated in different, private and public, collections in Europe and America, another reason that impedes the realisation of systematic studies.

In these plaques the artisans have introduced two important innovations in metal processing: first the intentional preparation of a copper and tin alloy, and second the sophisticated method of the lost wax casting. They are items of no more than 150 mm diameter. From a formal point of view these plaques were classified according to two variables (A.R. González 1998: 199; A.R. González et al. 2000): shape and decoration. Considering the first aspect, they were grouped as circular, rectangular and exceptional. Regarding the decoration of the plaques, three types were identified: "The personage of the empty hands", "The executioner or sacrificer" and "Two or more individuals". In all situations the central iconographic theme is an anthropomorphous figure in front view with its arms in flexion, a cephalic adornment and dressed up or ornamented with a tunica with echeloned drawings, spirals and rectilinear lines. In "The executioner" case, from its arms hang an axe, knife or representations of cut heads. Together with the central figure sometimes appear zoomorphous beings, such as felines, birds or saurians.

The distribution of this type of objects includes different places of Northwest Argentina including Chile and Bolivia. Discussing the different places of occurrence it was proposed that itinerant shamans might have distributed the artefacts as part of the political diffusion of the Aguada religious cult (A.R. González 1998: 100), but the centre of manufacturing is likely to have been Northwest Argentina (A.R. González 1992: 196).

**Precedent studies**

The number of technical studies of plaques of similar characteristics is very small. To our knowledge there are three publications on this theme: the first exhaustive work was published by Biloni et al. (1990). The study showed that the plaque investigated was made of bronze, with 2.5 weight % tin and less than 0.3 % arsenic. It was manufactured using the lost wax method (cf. Easby 1966: 74-76), polished and engraved. Lechtman (1991: 78) informed about the composition of another circular plaque cast in the lost wax mode. Its analysis gave 97 % copper, 1.31 % tin and minor quantities of iron, zinc, nickel, silver and lead. The last study was made by Scott (1998: 101-102) on a rectangular piece, again lost wax manufactured, whose chemical analysis gave: copper 88 %, arsenic 0.8 %, tin 8 %, antimony 2.7 %, and some traces of iron.

**Studies on three Aguada plaques**

The plaques which this work is concerned with were investigated using dimensional measurements, binocular stereoscopic observations, scanning electron microscopy and quantitative energy dispersive X-ray analysis. The compositional results were obtained as the average of a number of individual measurements. Not considered for the chemical composition were Na, Si, S, Cl, and Ca because they were assumed to be constituents of the patina of the plaques.

**Plaque N° 1 (Circular executioner)**

This piece, called the Bercheni plaque, was found in the Province of Salta (A.R. González et al. 2000). This disk is surrounded by a concentric band connected by five bridges with two appendices in the upper side (see Fig. 2). On the anterior face is an anthropomorphous central representation with a coiffure, fixed arms, and hands with three large fingers. From each arm hangs one instrument, in one case an axe and in the other a blade cutter. The legs end with three-toed feet. The surrounding band includes two saurian-formed animals, one of them without its upper jaw. The upper appendices end in ornithomorphous heads, with their eyes marked as a depression on the plaque. The back of the plaque is smooth, except behind the heads of the birds, where the anterior face follows their form.
The dimension of the plaque is 118.1 mm x 90 mm, with a weight of 115 g. Visual inspection determined the existence of white particles on the surface of the plaque with a tendency to concentrate in the grooves formed by the decorative design. Based on analogy to other objects from the same archaeological region it is assumed that the white substance was calcium phosphate, possibly a residue from the crucible or mould (cf. L.R. González 1997; L.R. González et al. 1999a). There are only minor fabrication defects. The mentioned absence of the jaw of one of the sauries is due to an imperfection in the mould during the pouring of the melted alloy. The border of the animal’s ear is rounded, suggesting a loss of fluidity of the alloy due to cooling. These details indicate that the entrance of the alloy into the mould occurred at the side opposite from the appendices with the birds figures. The composition was found to be 85.4 % copper and 14.6 % tin. The metallographic study revealed an as-cast solidification microstructure with a copper-tin eutectic in the interdendritic spaces and some globular oxides.

**Plaque No 2 (Rectangular executioner)**

The origin of this rectangular plaque is not known. On its anterior face there is an anthropomorphous figure whose head surpasses from the upper edge. The figure is represented in front view, with its two arms flexed. A knife, from its left arm hangs a knife and from the other an axe (Fig. 3). The feet are shown in side view. The head of the figure seems to be covered with a zoomorphous mask with a snout that stands out from the plaque plane. From the snout hangs a large moustache lying on the front of the plaque. In addition, the ears have circular depressions which probably held some ornamental stones.

The posterior face is smooth with a small depression that communicates to the snout. The dimensions are 94 mm in height and a weight of 135 g. The microscopic observations allowed us to identify a roughness of the surface, which we attribute to the used mould, the grinding substances and remains of a white powder which is supposed to have been used as a coating. The composition of the plaque is 96 % copper and 4 % tin. A complex substance on the surface was identified as paracatamite \([\text{Cu}_2(\text{OH})_3\text{Cl}]\), a well known corrosion product of copper and its alloys. Metallographic observation showed rounded grain boundaries with equiaxial tendency. The size of the grains suggested a low cooling process of the metal. Groups of slip lines indicate that the plaque undergone a light mechanical work.

**Plaque No 3 (Rectangular executioner)**

The plaque studied is of unknown origin and minor fabrication quality. The head is an anthropomorphic figure surpassing from the upper rim (see Fig. 4). The face of the person presents its eyes profoundly marked, the cavities may have sheltered ornamental or precious stones. A semicircular headgear surrounds the head. At both sides of the neck are easily seen holes that have been done by cold working. On the upper edge of the headgear, a cut indicates the position of the inlet of the mould.

The figure has flexed arms and from the right one hangs an axe. The leg lines disappear to the lower part and the feet appear only scarcely. The plaque weighs 53.5 g, is 81 mm in length, 51 mm in height, and has an average thickness of 1.7 mm. The thickness is less in the lower part of the plaque to the left, where a fracture is observed in an area of surface roughness. The lower left corner has a rounded edge. The chemical analysis indicates that this plaque is 93.5 % copper, 5.8 % tin and minor amounts of iron from the patina. The metallographic studies revealed a dendritic structure of copper and copper oxides, which tends to align along the mentioned fracture line. The plaque was made by the lost wax process, with the inlet from the mould lying on the central upper part of the plaque. A casting defect happened, resulting in an incomplete filling of the lower part of the mould.
but perhaps such evidence was removed by a different cleaning done by successive owners. The mentioned emulsion is white and could have been obtained from burnt and milled bones used as the very first fine cover of the wax model. This wax substance is commonly found in all the metallurgical products of Northwest Argentina during the subsequent periods. All the plaques show some fabrication defects always connected with an incomplete filling of the mould. Taking into account the tin quantities in the studied plaques and three others studied by different researches, we are able to confirm that the Aguada artisans worked in an authentic bronze. The concentrations of tin are different in the different plaques, but the incorporation of tin was deliberately done; in Northwest Argentina there is no associated minerals with both copper and tin and the registered compositions are too high to be explained by contamination. The alloy resulted from a technological choice, including the provision of different metals found in separate geographical areas. It also demonstrates an increase of the transformation operations and the capability put on them by artisans and other workers. In the Andes the interest to develop bronzes was an indication of the acceleration and demonstration of differential status and religious messages (Lechtman 1984: 45, 3988: 369). The use of tin means a change in the colour of metal, from red to golden. The golden colour remits to heliocentric cults that were fully developed in relation with the Aguada plaques (Pérez González 1986; A. R. González 1992; A. R. González et al. 2000). Moreover, the addition of tin lowers the melting temperature of copper and above all improves the filling conditions (Coghlan 1975; Craddock 1995; Tylecote 1979, 1987). A ltogether, very sophisticated considerations must have been taken into account in order to design and manufacture these difficult pieces.

The dissimilar content of tin in the composition of the plaques could have been a result of hazardous circumstances of their making. The considerably high percentage of tin observed in Plaque N° 1 suggests an intentional use of tin. One may hypothesise about the availability of tin ores. In Northwest Argentina, tin minerals are rare compared with copper ones, and the artisans of different geographical zones could have economised the use of tin, using the minimum quantities necessary in order to reach the intended valued conditions. Considering this point of view Plaque N° 1 may have been made by people with a significantly higher availability of tin, from ores or mercantilising.

The three studied plaques have cavities that may have been designed for precious or ornamental stones, as suggested by A.R. González (1992: 250). This ornamentation has not been found on the plaques. We suspect that this practice must have been more usual than commonly thought, and that the rare mention of ornamentation with known registered metal objects is due to the degradation of the natural adhesives used, and the subsequent loss of the inlays (L.R. González et al. 2001).

The laboratory studies emphasise the idea of the high symbolic value that these plaques may have had, considering the investment of labour, the technical knowledge involved, and the related degree of production organisation. Not only the paramount qualities of artist and artisans are detectable, but also the control over many physical and chemical properties connected with the material science applied during the manufacture of these objects.

The used ores may have been obtained from different mineral sources, which means a well-established knowledge of the regional geology and an efficient logistic organisation with its social and political legitimisation. The connected complexity inherent to the productive process suggests the involvement of specialised artisans who developed their task in an institutionally formalized frame. It seems clear that the technological style and the innovations that developed in the Aguada metallurgy did not respond to practical necessities, but to the development of dominant ideologies.

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References
A study of Hellenistic gilding practice and manufacture of funerary wreaths

Eleni Asderaki & Thilo Rehren

Abstract
A number of fragments from four Hellenistic wreaths were studied in order to better understand their manufacture and to identify suitable conservation treatment (Asderaki 2001). They were excavated during rescue work by the 13th Ephorate of Prehistoric and Classical Antiquities at the cemetery of ancient Demetrias in Magnesia, Central Greece. Three of the wreaths studied date to the early 3rd century BC, and one to the late 2nd century BC. Sampling was governed by the availability of fragments remaining from the conservation process, and analytical methods were chosen to provide as much insight as possible into the production and corrosion of these wreaths. In this paper, we concentrate on the gilding practices as well as manufacture techniques identified in the samples.

The wreaths were made to a high standard of craftsmanship, using often high quality material: ample gold leaf, cinnabar pigment and a pure kaolinite gesso. This use of high quality raw materials matches the relative scarcity of the wreaths among the overall number of tombs excavated: only about one percent yield remains of these ornamental items. However, despite their relative scarcity, they appear to have been made on a regular scale, using standardised methods and primary raw materials rather than merely recycling circulating metal and working on a semi-skilled ad-hoc level of craftsmanship.

Introduction
Gilded wreaths are relatively common items of personal adornment in Hellenistic graves, particularly in Macedonia proper (Robinson 1941; Makronas 1965; Despina 1980, 1996; Andronikos 1984; Vokotopoulou 1990). However, finds of Hellenistic wreaths spread from Thessaly in central Greece as far as southern Italy in the west (de Julius 1984: Cat. Nr. 20), Rhodes (Oddy et al. 1979) and Asia Minor in the east, and Cyprus in the south-east (Karageorgis 1973). The wreaths are generally imitating those made from a range of natural plants such as myrtle, oak, daphne, olive, etc. These particular wreaths studied here are representing myrtle, which is probably related to the worship of Demeter, Persephone and Aphrodite (Nikolaou, pers. com.), all goddesses adored in the area of ancient Demetrias and linked to the myrtle plant.

Despite this wide distribution, and the potential symbolic importance as a prestigious grave good, few technical studies are published concerning the manufacture of these, or indeed the type and quality of materials used to produce them. Here, a first attempt is made to rectify this situation.

General layout of a Hellenistic wreath
The majority of Hellenistic wreaths consist of a trephine as the central piece, which holds together the various attached decorative items. Probably, the trephine band stretched partly around the skull, with an opening in the front part above the forehead. According to the literature, a range of materials was used to build the trephine, including bone, wood, and metal. Of the four wreaths studied here, three had a leaden trephine, and one was made from wood. A tufted to the trephine were tufts comprised of leaves and fruits. The tufts were attached to the trephine in regular intervals of about two centimetres, fixed through holes punched into the soft trephine material. Each tuft consisted of a bunch of copper wires, which at their outer ends then either held the ceramic fruits or the copper-made leaves. A thin thread of organic fibre, wound around them and securing them safely, held the wires together (Fig. 1, see also Fig. 6). The majority of the individual items of the wreaths from ancient Demetrias were gilded with gold leaf, while only a few of the ceramic fruits were coloured by a red pigment. In the following, we will focus on the metal used to produce the wires, leaves and trephines, and the gilding technique applied in these wreaths.

Fig. 1. Sketch drawing of a linen thread securing a bunch of copper wires tightly together (drawing G. Kiassas).

Methodology and results
Since the major research questions in this study were concerned with the manufacture of the wreaths, the gilding techniques used, and the corrosion of the material, it was decided to use primarily image-based analytical methods. Although this required some sampling and sample preparation, it offered in return a detailed insight into the composition, treatment history and current situation of the metal. In addition, chemical analysis was done on the majority of the samples, using the already prepared metallographic mounts. Thus, the two main instruments used were the metallographic microscope and the Secondary Electron Microscope with attached Energy-Dispersive Spectrometer (SEM-EDS). In addition, some X-ray diffraction analyses were done to verify the nature of corrosion products. The samples were selected based on the ethical considerations necessary in any conservation and restoration work; thus, only tiny fragments, which were already separated from the main pieces, were sampled. This resulted in some limitations as to the extent of information gained, but this compromise was clearly necessary to minimize the adverse effect on the preservation of the archaeological material. The actual analyses then were done non-destructively, i.e. the mounted samples are still existent and available for any further investigation.

The metal
The wires and leaves were found to be both made of the same type of technically pure copper metal with a fair amount of copper sulphide inclusions throughout the body of the metal (see front page of this issue). In the majority of samples, the metal was totally corroded to form copper chloride, copper oxide and other corrosion products (see Asderaki 2001 for full details). Where preserved, this metal showed upon etching an annealed, recrystallised texture of equiaxed copper grains with no preferential orientation. The distribution and shape of the copper sulphide inclusions, however, were evidence of an earlier severe and directed deformation of the metal, most likely through hammering (Fig. 2). The copper leaves appear to have been cut into shape by chisels or scissors; in rare cases, hammer marks are still visible. Since the major research questions in this study were concerned with the manufacture of the wreaths, the gilding techniques used, and the corrosion of the material, it was decided to use primarily image-based analytical methods. Although this required some sampling and sample preparation, it offered in return a detailed insight into the composition, treatment history and current situation of the metal. In addition, chemical analysis was done on the majority of the samples, using the already prepared metallographic mounts. Thus, the two main instruments used were the metallographic microscope and the Secondary Electron Microscope with attached Energy-Dispersive Spectrometer (SEM-EDS). In addition, some X-ray diffraction analyses were done to verify the nature of corrosion products. The samples were selected based on the ethical considerations necessary in any conservation and restoration work; thus, only tiny fragments, which were already separated from the main pieces, were sampled. This resulted in some limitations as to the extent of information gained, but this compromise was clearly necessary to minimize the adverse effect on the preservation of the archaeological material. The actual analyses then were done non-destructively, i.e. the mounted samples are still existent and available for any further investigation.

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Fig. 2. Copper metal etched with alcoholic ferric chloride. Twinned crystals and elongated copper sulphide grains are clearly visible. Optical microscope, magnification 200x. Copper leaf from Tomb 393, 3rd century BC.
instances of good preservation of the metal texture in the corrosion products one can still see the characteristic distorted flow of the metal where it was cut.

Similarly, the wires were apparently made from copper metal hammered into a thin sheet and cut into strips, which were then hammered into a ‘G’-shaped wire (Fig. 3). This is in good accord with wire-making techniques throughout the pre-medieval world, and rules out the use of a drawing plate for their manufacture (Oddy 1987; Redfern 2000).

SEM-EDS analysis found no alloying elements in the copper metal, beside the before-mentioned ubiquitous sulphide inclusions. This indicates that the metal used was most likely not taken from circulating metal stock, which would probably have been dominated by recycled bronze scrap, and would possibly have been more highly oxidised due to repeated remelting. Instead, we assume that the metal was received directly from a primary smelting site, processing sulphidic copper ore. No further investigation towards the possible provenance of this metal was made. However, during the Hellenistic period, a number of copper sources in mainland Greece, especially in the Fthiotida area near Demetrias, were providing ample copper supplies to the urban centres of manufacture and consumption.

The metal strips of the lead trephines are 2 mm thick and 1.6 cm wide. Even though they are extensively corroded to cerussite, hydrocerussite and litharge (as determined by XRD analysis), they preserve enough metal in their core to judge their original composition. They were found to be made of almost pure lead. Optical microscopy, XRF and SEM-EDS analysis indicate the presence of some minor amounts of copper and silver, in the range of half a percent, as well as a trace amounts of arsenic and antimony. From these results, it appears that the metal was not subjected to desilvering (Rehren & Prange 1998) prior to its use here. The possible provenance of this metal was again not investigated; the most productive source of lead at that time, Laurion, is not far away from the area of ancient Demetrias, so it could be a possible origin of the used metal. Although very badly corroded, we can identify the shape and the way of their manufacture; the trephines were apparently made from lead metal hammered into thin sheets and cut into strips. Then holes were punched (Fig. 4) in regular intervals of about two centimetres, where the bunches of copper wire and leaves were attached. In a couple of samples, gilding is preserved. From optical microscopic observation we can assume that the gold leaf was applied straight to the metal surface either by burnishing it or by using a kind of glue to adhere it on the metal surface.

Gilding

Despite the badly corroded state of most samples, we were able to identify either by careful observation of the unmounted samples in the binocular microscope or – more rarely – in the mounted sections hidden underneath the corrosion layers remains of the original gilding. In the three wreaths from 3rd century BC graves, the gilding was always applied on top of a layer of siliceous material covering the metal core (Fig. 5). This gesso layer was identified by SEM-EDS analysis to consist either of pure kaolinitic material, indicated by the sole presence of alumina and silica in the EDS spectra, or as a calcareous clay, when the EDS spectra indicated in addition the presence of minor amounts of potash and calcium and iron oxide.

The gilding was found to be about one micrometer thick and to consist of technically pure gold; the levels of silver and copper were always found at or below the detection limit of the instrument, estimated to be in the range of half a percent in this heavy metal matrix. Thus, the gold was refined and parted prior to being hammered into gold leaf.
In the gilded copper leaf from the wreath from the 2nd century BC grave, the gilding was applied directly to the metal surface by burning the gold leaf straight to the clean metal surface. As in the other examples, the gold leaf was found to be about one micrometre thick and to consist of pure gold. The wire from this tomb, though, had the same gesso layer between the copper metal and the gold as the earlier samples.

**Non-metallic materials**

In addition to the various ornamental pieces made from metal, we identified ceramic beads and some organic fibres as integral parts of the wreaths. The beads came in two different sizes, and were identified to represent myrtle beads, matching the leaves. Some of them were gilded as well, and others were coated with a red pigment. In at least one case we were able to identify cinnabar as the red pigment. A with the gilded metal, both the gilded and the pigmented beads were first covered with a layer of gesso onto which the final surface decoration was then applied.

The copper wires which formed the tufts were best preserved near the holes in the lead trephines. Here, the corrosion of the copper had preserved among a group of three to four wires a central thread of organic fibre (Fig. 6), which apparently was both running parallel to the wires along their length, and then was wound round the lower ends of them, securing them by a knot. According to Sandra Bond and Liz Pye, both Institute of Archaeology UCL, these fibres are most likely from plants, not from animals, and may be linen. The tiny quantity of material preserved prevented any further research on this question.

**Conclusions**

Only about one percent of the 927 tombs found in the cemetery of ancient Demetrias preserve gilded wreaths made of composite materials. They represent the myrtle plant. From these wreaths four were selected for analysis, three of which dated to the early 3rd and one to the late 2nd century BC.

Several instrumental techniques were used to analyse minute fragments. The selection of the samples was made according to conservation codes of ethics, using only material which had already become detached from the major remains by corrosion.

The metal used for the manufacture of the leaves and wires was technically pure copper; the majority of this was corroded to form copper chloride, phosphate and oxide, malachite etc., preserving in many cases the ghost-like primary structure of the object. The residual metal after etching showed that it was hammered and annealed. The copper leaves have been cut into shape by chisels or scissors. To produce the wires, copper metal was hammered into thin sheet and cut into strips, which then where rolled or hammered to form a “G”-shaped wire.

Remains of the original gilding can be identified in most of the samples. In the wreaths of the 3rd century BC, gilding of the leaves and wires is invariably preserved on top of a gesso layer, which consists either of pure kaolinite or of calcareous clay. The gold leaf used is about one micrometre thick. It consists of pure gold containing less than half a percent of silver.

In the one leave studied of the wreath from the tomb dated to the 2nd century BC, the gilding was applied straight to the metal surface using the burning technique, while the wires had the same gesso layer as the earlier ones. Unfortunately, this being a unique sample, we can not identify whether this difference in the gilding technique represents a general change in craftsmanship from the early 3rd to the late 2nd century BC, or just a coincidental change in practice in this one piece. Clearly, further research is necessary here to answer this interesting question.

The lead trephines are made of almost pure lead which had probably not undergone any refining or desilvering. The lead metal was probably cast and hammered into sheets and then cut into strips, into which holes were punched so that the copper leaves and copper wires could be attached. The trephines were gilded as well. From all the above we see a consistently high standard of craftsmanship, which is indicated by the quality of the material used, such as the use of primary raw materials instead of recycled metal.

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In a recent paper in Paléorient, P. T. Craddock lays out his view of the development of the earliest metal smelting technologies in the eastern Mediterranean, proposing a path from crucible smelting in a hearth to furnace smelting. While much could be said about various parts of this synthesis of his thought on the topic, it was felt necessary to react to his presentation of evidence from two sites in the Southern Arabah Valley which are intimately linked with our work at these sites. He is a rather biased review of the evidence at these sites; this response attempts to give the proper data in their context. One might add that Craddock’s direct involvement in the fieldwork in this area was restricted to one season in 1976, when he (and his wife Brenda) came to our excavations at Timna as volunteers, and he was put in charge of the clearance of top soil in several small squares adjacent to Site F2, altogether lasting 6 days (6-11 October 1976).

According to Craddock (2001: 156), there is a coherent picture across the Mediterranean, southern Europe and the Middle East for crucible smelting of very rich copper ore, in a technique following on from the crucible melting of native copper. A gainst this essentially Chalcolithic smelting method, perceived and presented as universally valid, Craddock then contrasts our work at Timna, which presents a different approach to earliest copper smelting, based from the outset on an Egyptian New Kingdom tuyere. He ignores the fact that such general conclusions have been shown to be unacceptable by recent excavations at Chalcolithic A Bu Matar, in the Israeli Negev (Gilead et al. 1992: 12; Shugar 2000), where evidence for proper melting furnaces and tuyere fragments as well as crucibles exclusively used for melting/casting were identified. He also ignored the results of the classical smelting experiments by Tylecote (1974: 54; see also Rothenberg 1999:1): “One is forced to the conclusion that, while an area was used for copper metallurgy claimed to exist at Timna” is unfortunately not a discussion of the complete facts but a distorting presentation of the basic data of Timna Sites F2 and 39. The following paragraphs address some of the main points of this presentation.

Site F2. Craddock (2001: 156) begins his presentation of this site saying: “The excavation of F2 was supervised by the author of this review”. Craddock does not mention that there was a previous season of excavation earlier in 1976, directed by Rothenberg, which excavated and removed most of the early production centre of the site, including several typical prehistoric crushing mortars and a concentration of primitive slag (Rothenberg 1990: 6-9; Rothenberg & M erkel 1995; M erkel & Rothenberg 1999). This was the actual Site F2, where the pottery was found, which after very detailed petrographic and typological studies served as the archaeological evidence for the proposed Pottery Neolithic date of Site F2 (Rothenberg & Glass 1992; Rothenberg & M erkel 1995-96).

Craddock (2001: 156) continues: “The site lies in the general vicinity of much Late Bronze Age smelting activity and slag scatters are fairly common over the whole area”. This is not correct. Site F2 is located in the depths of the mining area of Timna, about 2 km from the nearest (New Kingdom) smelting site and not a single piece of slag was ever found in the mining area of Timna, except at Site F2.5 There is no “slag scatter anywhere in Timna outside the well-defined smelting camps of the Egyptian New Kingdom; all the earlier, pre-historical as well as Roman-Early Islamic smelting sites are located outside the actual Timna Valley (see the map in Rothenberg & M erkel 1995; Rothenberg 1990). Craddock (2001: 156) then goes on: “Most of the finds came from the surface or just a few cm beneath, and included a range of material from palaeolithic flints to modern cartridge cases.” This again is not correct: Site F2 was located on bedrock, on the surface of an almost flat slope, covered by a very thin (up to about 15 cm) layer of loose sand. Craddock’s description of the finds as a mixed “range of material” is also incorrect, since no Palaeolithic flint has ever been found in the A rabah or in Timna and certainly was not present in the findboxes from the excavation under Craddock’s supervision, which we have on record, kept by Craddock himself.6 We never heard about the find of “cartridge cases” in the Timna mines, hardly the right spot for hunting.

Craddock (2001: 156) then writes: “In the centre of the areas excavated was the only feature to survive... a small hollow containing ash in the bottom, surrounded by a setting of stones...”. It is important to keep in mind that the slags and other evidences of metallurgical activities were centred on this Late Bronze Age structure... It is very likely that the surficial slags and tuyere fragments (i.e. all the metallurgical remains at Site F2, B.R.) also belong to this operation... (i.e. melting of crushed slag during the New Kingdom). Very recently a TL determination of a piece of the slag has confirmed the Late Bronze Age dating...”. Craddock quotes as the source of this latter information a paper by H. Aupmann and Wagner “forthcoming “Prehistoric Copper Smelting at Timna” Thermoluminescence Dating of Slag from the Neolithic Smelter of Site F2. Levent”. Again, here are the facts: The small stone setting cleared by Craddock was not at all the centre of Site F2, but was located at the very end of the slope, about 30 m from the real production centre and major slag heap of the site (M erkel & Rothenberg 1999: 152, Fig. 6). Most of the finds of Site F2 were made at the early smelting site on top of the slope during my first season of work at the site in June 1976. According to the clear stratigraphy, recorded by Brenda Craddock, there was one stratum all over Site F2, the sand layer on bedrock, which contained the Neolithic remains. The stone setting excavated by Craddock at the lower part of the slope (I have intrusive, cut into here) was clearly intrusive, cut into the sand layer on bedrock (recorded by B. Craddock as Layers 1+2; L.1 representing the 1-2 cm loess on top of the sand) and was archaeologically obviously of a later date. Since both, the Late Neolithic smelting site uphill as well as the melting furnace of the New Kingdom on the lower slope, produced slag, we obviously have slags of two different dates at Site F2 and...
the radiocarbon and TL New Kingdom date of slag from Site F2 does not affect the Neolithic date of Site F2. As a director of the excavations at Timna, I discussed at the time the stratigraphy on site with Craddock and years later Craddock acknowledged the presence of earlier material at F2 in Leese et al. (1986: 117). Craddock’s current description of Site F2 in Palestine is unfortunately a misrepresentation of Site F2, presumably because our proposal of the Late Neolithic date of the site does not fit his theory of crucible smelting with blow-pipes as the universal rule for the beginning of extractive metallurgy in the entire Eastern Mediterranean and Middle East. The paper by Hauptsinn and Wagner mentioned above as forth running in Levant is also a strange story; I quote from a recent letter by Dr Kay Prag, editor of Levant: (late 2001). "I can assure you that no such article has been offered to Levant by any of the named parties, and that we do not have such an article, nor one which deals with the material from your Site F2 in Timna, in press in any form, or under consideration."

Site 39: Site 39 is one of the numerous pre- and proto-historic smelting sites located on low foothills along the mountain range of the Southern Arabah Valley, especially along its copper deposits. Site 39, like many of the other hill sites of the Arabah, had a workshop and/or habitation at the bottom of the hill (Site 39a), the actual smelting took place on the hill top above (Site 39b). Site a and b are connected by a short, well-trodden ancient path. A1 through looting we found numerous diagnostic flint objects as well as pot sherds dating to the Chalcolithic period. Crushed slag found in the workshop below was metallurgical the same as the smelting slag around the furnace on top of the hill (M erkel & Rothenberg 1999: 161-2) and it is obvious that we are dealing with one site of one date. Craddock (2001: 156) begins his assessment of this site in saying: "For many years the dating was based on the flint typology of the inconvenient radiocarbon date of 1945 ± 309 BP BM-1 116 (Burleigh & Hewson 1979), calibrated (INTCAL 98) to lie between 800 BC and 700 AD at 95.4 % confidence, being ignored. The sample was of charcoal taken from the furnace wall itself and submitted by Barbetti and Rothenberg". Here are the facts: This particular sample was taken by Barbetti (contrary to Craddock’s statement above, without my knowledge3) from the furnace at Site 39b which had been backfilled by us with earth from down the wadi in order to prevent the collapse of the furnace excavated by us. Barbetti did not know about this and the charcoal sample came from this backfilled material (as already published in iams 10/11, 1987: 14-15). It is Craddock who ignored the important note by Burleigh & Hewson (1979): "BM 1116 is also apparently invalidated by missassocation."

Craddock then opposes the new radiocarbon date - OXA 7632 5485 ± 45 BP calibrated to 4460-4240 BC at 95.4 % confidence- of a sample from the kitchen of Site 39a (supplied by B.R.), which fits well our Chalcolithic dating of Site 39a + b by archaeological and metallurgical evidence.4 He is ‘explanation’ "that all the enclosures (i.e. habitations, workshop and upper stone structure of the actual smelting furnace, B.R.) are Chalcolithic, and may have been associated with metallurgy at the time, but the furnace and slag remains are of a much later period" (Craddock 2001: 156), does not really make archaeological or metallurgical sense. To conclude, Craddock in his discussion of the earliest phases of extractive metallurgy in the Middle East attempts to draft a coherent theory of uniform technological progress across the vast range of cultural and ecological parameters controlling the regional development in the Levant, making eclectic, disturbing use of data from Timna to fit his theory. This is in surprising contrast to his discussion of the evidence for Bronze Age smelting furnaces later in his paper (Craddock 2001: 161-2), where he explicitly acknowledges the technological difficulties in smelting copper in crucibles, and concludes: ‘The answer was to develop the furnace as a specific structure for the smelting of metals. From the remains already published…..[it] seems that there was a variety of furnace types in use through the eastern Mediterranean and Middle East. This variety suggests local development, …, but must also to some degree reflect local materials and conditions.” It appears odd to me that the much earlier phases of incipient extractive metallurgy should have been so well understood with such opprobrium for crucible smelting that the much earlier phases of incipient extractive metallurgy at the site were recorded by the area supervisor, an experienced archaeologist, as ‘furnace lining’. Only subsequent studies in the laboratory showed that these lumps could have been used as a kind of tuyere.

3 Rothenberg and Merkel, 1995: 3. The only exception to this statement is a small concentration of slag, Site N3, probably of Chalcolithic date, found at the eastern edge of the mining area, but no evidence for smelting was found at this site. Segal, Rothenberg and Bar-Matthews, 1998.

4 Following Craddock’s paper, we checked once more the findboxes of this excavation, and nothing of such “range of material” was found or recorded.

5 Cf. section drawing by B renda Craddock in M erkel and Rothenberg 1999: 153. A dating according to Craddock’s Site Notebook (1976: 30) the TL sample and charcoal samples for C14 were taken by him from Layer 3 of Square 3. i.e. from inside the New Kingdom melting installation.


7 Bercovic in Rothenberg 1978: 16-20. S. Rosen, the prehistorian of Ben-Gurion University, Beersheba, has lately concluded the re-investigation of theflints from Site 39 a+b and dated this material to an early phase of the Chalcolithic period (forthcoming in the new edition of Rothenberg 1978.)

8 Cf. an appeal to the editor of Radiocarbon, in iams 10/11, 1987: 14-15, which also relates to the radiocarbon sample taken by Craddock at Site F2.

9 Cf. an appeal to the editor of Radiocarbon, in iams 10/11, 1987: 14-15, which also relates to the radiocarbon sample taken by Craddock at Site F2.

References


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