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SHAPING BRONZE BY HEAT AND HAMMER: AN EXPERIMENTAL REPRODUCTION OF MINOAN COPPER ALLOY FORMING TECHNIQUES

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ABSTRACT

The compositions of copper-base tools, weapons, ornaments and ceremonial metalwork from numerous Late Bronze Age Aegean sites reveal a pattern of specific alloy combinations for the fabrication of certain classes of objects. Thus the majority of weapons and tools were made of high tin bronze whereas bronze statuettes, tripods and cauldrons contain small amounts of lead and in some cases tin is present in low amounts. Such diversity reflects the direct relationship between the compositions of prehistoric bronze objects and the art of their fabrication, because both the alloy additions and the impurities exert a pronounced effect on the forming capacity of alloys. In order to understand the correlation between composition and formability of Minoan bronzes, replica compositions with varying tin and lead contents were experimentally reproduced and their forming capacities were tested. Deformation and heat treatment of five tin and two leaded tin bronze alloys was attempted in order to replicate the forming stages for the shaping of cutting tools and bronze sheet for vessels and cauldrons. The amount of cold-working and annealing intervals, required to test the effects of workability on alloy properties, has been reflected as hardness values and transformations of the structural characteristics for each sample. It has been shown through the course of the experiment that high tin bronzes could be formed by frequent, short annealing stages at 600˚C and that it is possible to work-harden leaded bronze as long as time and temperatures are closely monitored.

KEYWORDS: annealing, cold-working, ductility, hardness, leaded-bronze, recrystallisation

1. INTRODUCTION

The production of copper alloy artefacts that gradually replaced unalloyed metalwork of the Final Neolithic-Early Bronze Age cultures of Anatolia and Europe was an innovation that spread rapidly across the Aegean in the beginning of the 3rd millennium BC (Roberts 2009; Muhly 2008; Doonan and Day 2007; Primas 2002). The early methods of bronze working have been often described as complex sequences with stages dependent upon the nature of the material and the skilful actions and choices of the bronze smith during manufacture (Thornton 2007; Muhly 2002; Ottaway 2001). Two main techniques, namely casting and fabrication by direct shaping were commonly used in prehistoric foundries for the production of copper alloy artefacts. Given that the physical properties of any alloy determine the degree to which it can be worked, it has been suggested that ancient metalworkers utilised specific compositions for the manufacture of certain types of artefacts (Kienlin and Pernicka 2009; Budd *et al* 1994; Northover 1989; Craddock 1976).

Ranging alloying additions to copper give rise to distinct properties that could either enhance or reduce workability and it has been argued by researchers that prehistoric metalworkers chose specific alloys based on the intended shape, form and function of the object under construction (Craddock 1995; Dungworth 1996; Tylecote 1987). This pattern is obvious from the early stages of the Bronze Age in Europe and the Mediterranean but becomes more apparent towards the Late Bronze Age when tin becomes widespread and circulation in metals rises dramatically (Muhly 1999; Nakou 1995; Pernicka *et al* 2003). An equally important factor influencing the finished product's form involves the fabrication mode, dictated by the craftsman's actions which have been often overlooked. Applications of metallographic analysis on archaeological material have enabled us to promote a better understanding of the various stages of bronze working (Scott 1991). Further experimental approaches provided valuable insights to the experiential side of working with bronzes, manipulating the material and being faced with real problems that need to be addressed accordingly (Piccardo *et al* 2010; Heeb 2009; Andrews and Doonan 2005).

The current paper discusses the results from experimental reproductions of forming methods for certain bronzes in order to investigate the correlation between composition and microstructure of the finished product through the process of fabrication. The experiment was designed in order to replicate the possible ways of working copper alloys that were common during the Late Bronze Age in Minoan Crete and the Aegean islands and therefore tin and leaded tin bronzes have been used. More specifically various stages of work hardening by cold-working and annealing were attempted in order to quantify the extent to which deformation and recovery were undertaken in prehistory and untangle crucial choices made by the craftsmen through each step of the metalworking process *(chaîne opératoire)*.

2. FORMING TECHNIQUES OF THE MINOAN BRONZE INDUSTRY

Chemical analyses undertaken in the past have indicated that during the Late Bronze Age a common metals technology was in use at major Minoan centres such as Knossos, Phaistos, Aghia Triada, Gournia and Palaikastro (Craddock 1976; Evely 1995; Mangou and Ioannou 1998). The percentage of tin varied in the narrow range of 5-8% and generally other elements such as As, Sb, Ni, Fe were at the trace level (Mangou and Ioannou 1998, 99). Metallographic investigations on such material revealed how cold working, annealing and forging were the dominant methods for the fabrication of weapons and tools (Tselios 2008). In most cases a final hammering stage followed annealing but there are examples of artefacts indicating the application of quite diverse treatments. For instance Betancourt *et al* (1978) found that two objects from Gournia, a strip and a needle, were both cast, cold-worked and annealed, but with no indication of a final cold-working stage. Northover and Evely (1995) showed that their sampled tools had been worked after casting by combining cold forging with annealing to ensure toughness. Metallographic examination of Minoan weapons and personal objects at the Ashmolean Museum revealed how the metalworkers were competent in working all types of alloys for fashioning various classes of objects (Baboula and Northover 1999).

The bronze tools and weapons from Knossos contain higher tin contents than the corresponding vessels but there are cases excluded from this general pattern. For example whilst knives, spearheads and double edged axes contain 9-11% tin, the chisels and swords show average contents of 3-5% and the adzes and hammers strangely enough contain lower tin levels (0.21-0.22%). The vessels in general do not exceed 6% tin except for the case of a pan containing 10.34% (Mangou and Ioannou 1998). Lead contents are generally low in all classes of objects ranging between 0.01-0.07% with the exception of four double edged axes, a chisel and two pitchers with lead contents close to 1%. The manufacturing process to create these pitchers might have involved the production of thin sheets of bronze which were joined to form the walls of the vessels. Based on the above archaeological examples the experiment was designed to reproduce specific fabrication sequences on bronze with similar compositions attempting to replicate and understand the actions of prehistoric smiths.

3. METHODOLOGY OF THE EXPERIMENT

Four tin bronze and three leaded-bronze alloys were used for the experiment with compositions as follows: 2%Sn, 6%Sn, 10%Sn, 15%Sn and 6%Sn + 10%Pb, 6%Sn + 2%Pb, 10%Sn + 2%Pb. The samples, roughly cylindrical in shape and approximately 9mm in thickness, were cut off from feeders of previous casting experiments with sand moulds (Ottaway and Seibel 1998). The working stages could be summarised as follows: treatment A involved cold-working of 40% reduction in thickness, annealing at 500˚C for 10 minutes and 20 minutes intervals and a second cold-working phase of 40% reduction. Treatment B started with a cold-working phase of 26.6% reduction in thickness followed by annealing intervals at 500˚C for 10 and 20 minutes and continued with a second deformation phase for a further 26.6% reduction in thickness. A second annealing phase followed (500˚C for 10 and 20 minutes) and a final reduction of 26.6% by cold-working. Treatment C consisted of two cold-working stages identical to Treatment A except for the annealing temperature which was raised to 600˚C in order to compare the extent of recrystallisation with varying time (table 1).

In an attempt to replicate closely prehistoric conditions, cold-working was achieved by direct hammering on an anvil rather than being undertaken mechanically in the rolling mills. Annealing conditions were chosen on the basis of experimental work undertaken in the past and the samples were processed in an annealing oven (Budd 1991a; Budd and Ottaway 1991; Papadimitriou *et al* 1996). The relatively low temperatures of 500 and 600˚C and the short lengths of 10 and 20 minutes are sufficient for partial recrystallisation, which recovers ductility. Higher temperatures and longer times were avoided as they lead to homogenisation. Previous experimental work has shown that copper-tin alloys annealed at 600˚C still show signs of coring while at 700˚C they become completely homogenised (Budd 1991b; Dungworth *et al* 1999).

4. **MECHANICAL OBSERVATIONS AND MICROSTRUCTURAL FEATURES**

During hammering the rate of cracking was monitored in order to compare variation based on composition and mode of working. Samples that were processed by treatment A sustained the first cold-working phase without cracking. After annealing, a second cold-working took place during which samples of low tin contents (2%, 6%) were successfully reduced to nearly 80% without cracking. Samples with 10 and 15% tin (A.10.00.10, A.10.00.20, A.15.00.10 and A.15.00.20) fractured at reductions lower than 80% as shown in Table 1. The same is true for the six leaded bronze samples and it could be said that the annealing conditions of treatment A were not sufficient for recovering ductility of the high tin and leaded samples.

Treatment B involved a cycle of three hammering stages separated by two annealing intervals and appears to be closer to Minoan practices as indicated by recent findings (Tselios 2008). After the first cold-working none of the samples had signs of cracking or fracturing. They were all reduced close to the intended figure (26.6%) with a range between 22-28%. The second cold-working has been successful at reducing the samples in the range of 50-58% without cracking except in the case of the leaded 6% tin sample (B.06.10.20). The sample contains substantial amounts of lead (10%) and was annealed for 20 minutes. Its equivalent B.06.10.10 did not crack even though it was annealed for only 10 minutes but it should be noted that difference in percentage of reduction between the two (54.79 and 56.60% respectively) is an equally important determining factor. The third cold-working stage caused most of the samples to fracture except those containing 2% and 6% tin. The majority of the samples cracked before reaching 80% of total reduction and the average reduction at which fracture occurred is around 77%. Therefore it could be said that treatment B, i.e. gentle reduction in thickness with frequent annealing, has been more effective than treatment A for working bronzes containing up to 10% tin. Metallographic examination has shown how the δ phase sustained severe deformation through hammering but did not cause cracking due to a second

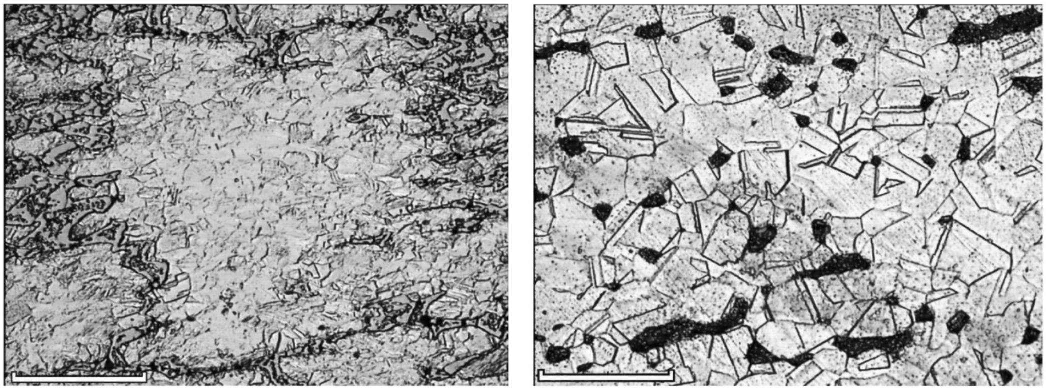
1.a 1.b

Figure 1: a) High tin bonze hammered and annealed at 500°C showing nearly complete recrystallisation with some strain lines still evident. The extensive bluish areas are rich in the $\alpha + \delta$ eutectoid (scale bar 0.2 mm). b) High leaded-bronze with fully recrystallised grain structure after annealing at 600°C. The grains are large and annealing twins are common. Black areas are segregated lead inclusions (scale bar 0.1 mm).

annealing episode (Figure 1a and Table 1).

Treatment C was designed to test the variable of temperature affecting workability. Conditions were altered by raising the temperature to 600˚C while keeping annealing time at 10 and 20 minutes in order for the results to be comparable. Table 1 shows that although most samples were reduced above 40% the tin bronzes sustained deformation without failing while the leaded alloys were prone to cracking. The annealing stages at 600˚C proved to be ideal for strain relief and partial recrystallisation that recovered ductility to a greater extent than at 500˚C. This is also evident from an examination of the metallographic sections (Figure 1b). Therefore during the second cold-working phase compositions of up to 10% tin were successfully hammered close to 80% reduction while the 15% tin samples shown only minor cracks. Conversely all leaded samples fractured at around 75% reduction showing that annealing at 600˚C proved more efficient for working such alloys.

5. EFFECTS OF FORMING TECHNIQUES ON ALLOY HARDNESS

In the current study differential increase in hardness was affected by the ranging conditions of each working treatment (variables of temperature, length of annealing, final reduction) and alloy composition. The hardening performance of the tin bronzes through each treatment has been measured by the Vickers Hardness test and is graphically presented in two diagrams. Figure 2 represents hardness values per composition for those samples annealed for 10 minutes. The curve for treatment A shows that hardness rises proportionately with tin contents up to 10%Sn while at 15%Sn hardness decreases by around 30HV. Treatment B had a better performance as it yielded high values for the low tin alloys and a constant rise in hardness at higher tin contents. Treatment C yielded the lowest value for the 2% alloy and the highest for the 15% alloy (300HV). Apparently treatment C has been proven slightly more efficient than treatment B as it promoted increase in hardness at a constant rate.

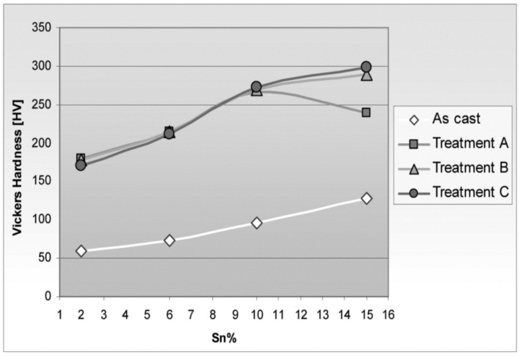

Figure 2: Hardness data of the Cu-Sn alloys. The basal curve represents hardness of the as cast samples and the remaining, hardness achieved through each working treatment (annealing time 10min).

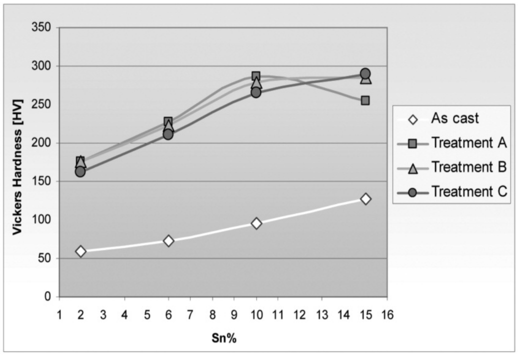

Figure 3: Hardness data of the Cu-Sn alloys. The basal curve represents hardness of the as cast samples and the remaining, hardness achieved through each working treatment (annealing time 20min).

As shown in figure 3 (annealing time 20 min) treatment A remains predominantly more efficient for compositions up to 10%Sn but drops considerably at 15%Sn. During treatment B as tin contents increase from 2 to 10% there is a stable rise in hardness which continues, albeit at a reduced rate towards 15%. Although treatment C produced the lowest hardness values for low tin contents it followed the most pronounced increase curve.

An evaluation of the impact of lead on hardness values was attempted through a comparison of the following three compositions: 6%Sn, 6%Sn+2%Pb, 6%Sn+10%Pb. As could be deduced from the diagram in figure 4 (annealing time 10min) there is a slight increase in hardness from 0 to 2%Pb and a considerable decrease from 2 to 10%Pb which is prevalent for all treatments. Samples in the as cast condition follow the same general trend even though there is no pronounced decrease between 2 and 10%Pb contents. The same general pattern was observed for samples annealed for 20 minutes (Figure 5) where the rise in hardness from 0-2%Pb during treatment C is minimal and almost absent during treatments A and B. The 10%Pb samples display considerably lower hardness values than those containing 2%Pb.

The general outline of each treatment performance was evaluated in terms of efficiency and is presented in diagram form (Figure 6). It should be noted that annealing eliminates slip bands caused during deformation and therefore higher temperatures and/or longer intervals in the oven generally restore ductility and result in reduced hardness. The current experimental results however show two apparent anomalies. After treatment A the 10% bronze is harder than the 15% bronze and annealing for 20 minutes left the 10% bronze harder than if it had been only annealed for 10 minutes. This discrepancy might be explained by the fact that the majority of dislocations and strain energy are not entirely removed by usual recovery treatments, which means that properties such as hardness are not always severely affected (Guy 1960). Additionally previous experiments have shown that annealing tin-bronzes between 400 and 600˚C for 30 minutes paradoxically resulted in slightly increased hardness values than those noted for shorter periods but only in rare instances (Dungwoth et al 1999). In any case

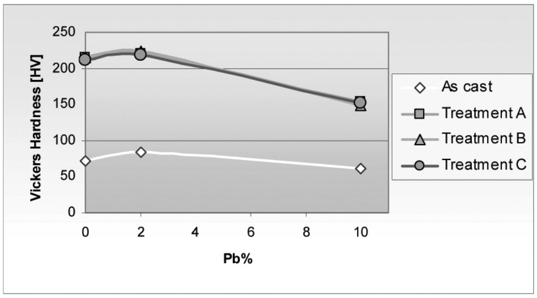

Figure 4: Hardness of Cu-Sn-Pb alloys. Stable tin content (6%) and varying lead contents (0, 2, 10%) (annealing time 10min)

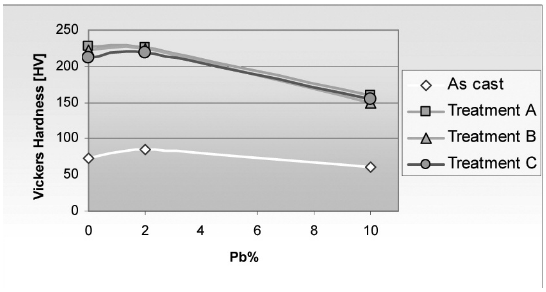

Figure 5: Hardness of Cu-Sn-Pb alloys. Stable tin content (6%) and varying lead contents (0, 2, 10%) (annealing time 20min)

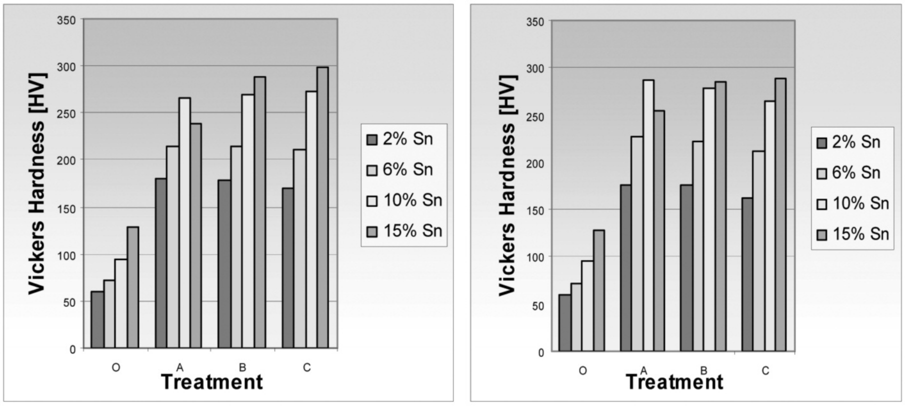

Figure 6: Hardness of the Cu-Sn alloys. The maximum hardness values achieved through each treatment could be compared. (a. ann. time 10min, b. ann. time 20min)

such ambiguous data should be explained due to the effects of homogenization and any associated remnant coring.

Treatment A appears more efficient for low tin contents as evidenced by the high hardness values whereas it appears inappropriate for 15%Sn alloys. Treatment B produced almost as high values as treatment A for low tin contents and significantly higher values for 15%Sn contents. Therefore three hammerings, intervened by two annealing stages appear suitable for working a range of compositions in tin bronzes. Treatment C appears inappropriate for low tin contents and more auspicious for higher levels of tin. Overall the short annealing intervals of 10 minutes were proven suitable for hardness increase whilst the 20 minutes intervals have been clearly more efficient for increasing the hardness of the 10%Sn samples (Table 1).

6. CONCLUSIONS

Prehistoric metalwork was commonly produced through certain forming techniques employing specific alloys for certain classes of objects. In order to understand the correlation between composition and formability of bronzes used in Crete during the Late Bronze Age ancient compositions with varying tin and lead contents were reproduced while their forming characteristics and enhancement of physical properties were experimentally tested.

During deformation of the low tin bronzes, micro-cracks appeared inside the eutectic especially for the final reductions attempted (80%) but all samples were worked relatively easily. Therefore a treatment which involves as less as an initial cold-working, one annealing stage and a second cold-working appears efficient for working low tin bronzes that were in use for vessels. A lengthy process with numerous reduction stages is not necessary for the manufacture of objects from low tin bronze and might have been avoided in the past. In the alloys with 10-15% tin, cracks appear across the thickness of the samples for deformations above 50%. The 15% tin alloys present wide transverse cracks rising to the surface and hindering formability. It has been shown that the hard eutectic in such alloys could be limited by frequent annealing at 600°C for as short a period as 20 minutes. It follows that in prehistory high tin bronzes for edged tools or weapons were most probably worked by hammering at room temperature and heat treatments similar to the one performed.

The presence of lead in copper alloys diminishes formability by causing cracks and surface defects. Lead causes work hardening at the very early stages of deformation. As shown during the course of the experiment low lead contents (6%Sn+2%Pb) give rise to greater hardness than pure tin bronze (6%Sn), while increased levels of tin significantly reduce hardness. At comparable annealing conditions, the high leaded alloys reached more pronounced recrystallisation and display lower hardness and higher ductility than the pure tin bronzes. Although it is possible to work-harden leaded bronze it should be noted that such alloys were usually utilised for the production of cast rather than wrought objects in prehistory (Hughes et al 1982: Papadimitriou 2008).

By following simple stages of repeatable actions for working with bronzes, sequential steps of the operational chain for artefact manufacturing were reconstructed. In each step the most likely variables that would influence a craftsman's choice were determined and it was concluded that combinations of cold-working and short annealing stages are effective for shaping various objects from a range of alloy compositions. Clearly there are numerous ways by which metal artefacts could be fabricated but what this paper aimed for was to display some of the potential manufacturing methods and the inherent intricacies the prehistoric bronze smiths had to cope with. Taking into consideration the diversity of skills and experience necessary for the effective manipulation of metals and in turn the difficulty we face to replicate closely such practice by modern means this experiment serves to boldly illustrate the surprising complexity behind the ancient

Treatment A	1ST CW Red. %	2ND CW Red. %	Treatment B	1ST CW Red. %	2ND CW Red. %	3RD CW Red. %	Treatment C	1ST CW Red. %	2ND CW Red. %
A.02.00.10	41.85	81.25	B.02.00.10	23.27	52.57	78.69	C.02.00.10	42.75	79.99
A.02.00.20	42.33	81.76	B.02.00.20	22.71	58.76	80.35	C.02.00.20	38.88	82.21
A.06.00.10	37.77	78.33	B.06.00.10	24.47	50.53	80.85	C.06.00.10	41.76	78.82
A.06.00.20	43.52	78.82	B.06.00.20	22.93	52.28	79.79	C.06.00.20	41.00	76.50
A.10.00.10	37.14	**72.85**	B.10.00.10	21.28	50.54	**70.87**	C.10.00.10	44.73	78.94
A.10.00.20	41.76	**77.05**	B.10.00.20	22.34	52.13	79.80	C.10.00.20	44.73	78.94
A.15.00.10	39.40	**67.64**	B.15.00.10	23.40	51.06	**80.85**	C.15.00.10	45.25	**81.57**
A.15.00.20	39.40	**51.76**	B.15.00.20	24.38	54.07	**75.36**	C.15.00.20	38.33	**76.66**
A.06.10.10	39.99	**78.94**	B.06.10.10	24.47	54.79	**70.87**	C.06.10.10	**44.73**	**85.78**
A.06.10.20	43.50	**70.00**	B.06.10.20	23.53	**56.60**	**79.79**	C.06.10.20	**44.73**	**77.89**
A.06.02.10	41.10	**66.66**	B.06.02.10	29.15	52.10	**81.29**	C.06.02.10	37.50	**72.50**
A.06.02.20	40.94	**67.61**	B.06.02.20	28.57	55.61	**80.27**	C.06.02.20	40.92	**79.00**
A.10.02.10	49.40	**68.81**	B.10.02.10	23.40	51.07	**76.12**	C.10.02.10	44.73	**65.78**
A.10.02.20	38.33	**72.21**	B.10.02.20	24.62	57.70	**81.90**	C.10.02.20	**43.80**	**76.64**

Table 1: Percentage of reduction in thickness through treatment A, B and C (each sample is represented by: Treatment - Sn%- Pb%- Annealing time. Figures in bold underlined represent fractured samples)

craft of the bronze smith.

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