Ingots and the Bronze Age Copper Trade in the Mediterranean: a progress report

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Acknowledgments
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The last twenty years have seen an increase in scientific studies of archaeological materials resulting from the desire for greater precision in archaeological data. Research on ancient metal objects has contributed significantly to the data, due to a growing scientific interest in ancient materials and the application of new techniques of metallurgical analysis to metal artifacts. Modern metallurgical studies have honorable predecessors in C. H. Desch and the Sumerian Copper Committee who were active in the 1920's (see Witter 1938). This group, on behalf of the British Museum, attempted to locate the sources of metals used in ancient Mesopotamia through wet chemical analysis of elements present in artifacts and ores. This work was generally reliable and, although wet chemical analysis has been largely replaced by emission spectroscopy and X-ray fluorescence, may still be cited with some confidence.

Most recent programs of ancient metallurgical research have originated in Europe and the United Kingdom. One of the major groups working in this area is based in Stuttgart (Junghans *et al*, 1968); their main interest is in European copper sources and artifacts. Pittioni and his colleagues in Vienna have made detailed studies of the prehistoric copper industry in Austria (see Pittioni 1959). Determination of the products of various copper sources by emission spectroscopy has not always been consonant with archaeological data, a common problem when scientists work without the collaboration of archaeologists and historians. In the United Kingdom, the major research project is that of Hugh McKerrell of the National Museum of Antiquities of Scotland. Among many important projects, McKerrell has performed analyses of hundreds of metal objects in southwestern Asia, using a portable X-ray fluorescence unit. The analyses have been computerized and are being turned over to excavators and museum officials. Some of the significance of this large body of material is lost since the analyses are presented as a gross unit including all of Southwest Asia, with only rough chronological distinctions preserved. The dating system used is that proposed by excavators of the material or written in the museum catalogue with no integration of dates into a related body. McKerrell has not concentrated on trace element analysis and questions of provenance. His major emphasis is on the reconstruction of the development of alloying techniques, especially the widespread use of arsenical copper in the third millennium B.C. In the United States, George Rapp and Strathmore Cooke at the University of Minnesota and Eiler Henrickson of Carleton College are computerizing the data from neutron activation analyses of native coppers from all parts of the world. They hope to identify copper sources and relate them to artifacts produced in the Bronze Age.

Archaeologists and historians have explored ancient metallurgy from their own perspectives. Branigan (1974) presents a large body of archaeological material drawn from Crete, but uses little scientific data and makes some misstatements about scientific aspects of metallurgy. Ufuk Esin (1967) has had analyses of hundreds of Turkish objects performed by the laboratory in Stuttgart; it is useful to have these analyses presented in one place, but the accompanying commentary is not so detailed as it might have been. Muhly's discussion (1973) of copper and tin and their trade in the Bronze Age concentrates on philological and historical questions and, while it does not use scientific analyses, does provide a background for scientific studies. Numerous other archaeological studies involve single sites or areas, or single classes of objects, most notably weapons. Deshayes' monumental classification of bronze objects (1960) deserves attention partly because of the amount of material presented and the fact that the data are computerized for efficient retrieval.

Discussion of these publications indicates the need for integration of the scientific and archaeological aspects of research in order to produce more meaningful data for reconstructing technological history. Since we are an archaeologist, a metallurgist and an historian, we hope to develop a pattern of collaboration which can cover all aspects of ancient metallurgical problems. Such collaboration will enable us to extract the maximum amount of information from scientific data, and to understand the potentials and limitations of scientific equipment and data so that they can be effectively exploited. Our efforts are encouraged by the facilities available to us at the University of Pennsylvania—the University Museum with its rich collections and active excavators, the Museum's Applied Science Center for Archaeology (MASCA) which has pioneered in the application of science to archaeological problems, the Laboratory for Research on the Structure of Matter with its comprehensive range of scientific equipment and scientists, and geologists involved in research in the Mediterranean area.

Copper ingots have been selected as the vehicle for the first stages of our collaborative research. In the third and second millennia B.C., copper was the metal most commonly used in the Mediterranean area and the Near East, although after ancient smiths developed the technique of alloying copper with small amounts of arsenic and later of tin it was seldom used alone. Trade in the components of the resulting alloy, bronze, produced by the mid-third millennium B.C. the first great period of international contact. Ingots are a primary form in which copper was cast for shipment and therefore a single ingot was probably made from the ores of a single mine. There should be no confusion in ore sources which one expects in the case of artifacts, many of which were fabricated with varying amounts of scrap metal. Wide distribution of ingots too is significant; they are found in Israel, Cyprus, Turkey, Crete, Greece, Sicily and Sardinia.


1

The shapes of the ingots may reflect the metallurgical techniques used in their production. Bun or plano-convex ingots may have been formed in the bottom of the furnace in which they were smelted, with destruction of the furnace necessary to remove the ingot. Such ingots are most frequent in the Near East, while oxhide ingots are characteristic of the Mediterranean copper trade. The latter would have been cast, perhaps in a pit, to form their ear-shaped handles. Bar ingots from Israel probably are a secondary form, cast from the metal of a larger ingot to facilitate production of a particular object, perhaps a sword or dagger. Thus the techniques of production may have been conditioned by the location of manufacture and the immediate use, either object production or shipment, to which the ingot was to be put.

Because a number of ingots have been found on Crete—at Tylissos, Hagia Triada, Kato Zakro and Mochlos—and people identified as Cretans are shown bearing ingots in Egyptian tomb paintings, Crete has been called the major source of oxhide ingots and an exporter of copper in the Bronze Age (Merrillees 1974). Alleged deposits of copper on the island are cited as the source of the metals used to produce the ingots and objects of local manufacture (Faure 1966). In order to verify the reports of copper deposits, Muhly and George Rapp surveyed in Crete during the summer of 1974. A number of the copper deposits discussed in the literature are represented only by green stains on rocks and some are serpentine rather than copper. Copper is present at Miamou and Zervos, near Lentas in southern Crete, but the extent of the deposits and the dates of their use are not certain. The Miamou deposits deserve a thorough investigation because of their proximity to the Minoan sites of Phaistos, Hagia Triada, Lebena and the possible port at Komo. A mine at nearby Chrysostomos yielded a very low grade copper in the 1950's, making it unlikely to have been significant in Minoan metallurgy. One dubious copper mine in western Crete south of Khania was discovered; the mine had been destroyed by bulldozers during road building, but locals reported that it had been worked in the early years of this century. The results of the survey indicate that Crete should probably not be called a major producer of copper in the Bronze Age.

Cyprus, on the other hand, does have geologically verified and extensively exploited deposits of copper (Bear 1963). Several studies have been made of ancient mining on the island; these deal chiefly with the Roman period (Rickard 1930; Davies 1932). Copper working on Cyprus is documented for the Late Bronze Age by the presence of implements and slags at Apliki (Taylor 1952), Enkomi (Dikaios 1971), Athienou (Dothan and Ben-Tor 1974) and Kition (Karageorghis 1973:10), and its history pushed back to the Early Bronze Age on the basis of finds at Ambelikou (Dikaios 1946). The abundance of copper and the evidence for its working on Cyprus make it unlikely that Bronze Age metal workers imported the metal, either smelted or raw, from elsewhere. H. G. Buchholz (1967) has attempted to identify Cypriote ores on the basis of their trace element content determined by emission spectroscopy; objects with zinc and no tin are called Cypriote. Zinc may rather be an indication that sulphide ores were used and tin was probably added delib-

1
Map showing distribution of copper oxhide ingots. After map published in H. G. Buchholz and V. Karageorghis, *Prehistoric Greece and Cyprus, An Archaeological Survey*, London (Phaidon Press), 1973, p. 61, fig. 26.

2
Bun or plano-convex ingots from the Arkalokhori Cave, Crete, as displayed in the Heraklion Museum. Excavated by the late Spyridon Marinatos. The ingots are probably to be dated in the 16th century B.C.

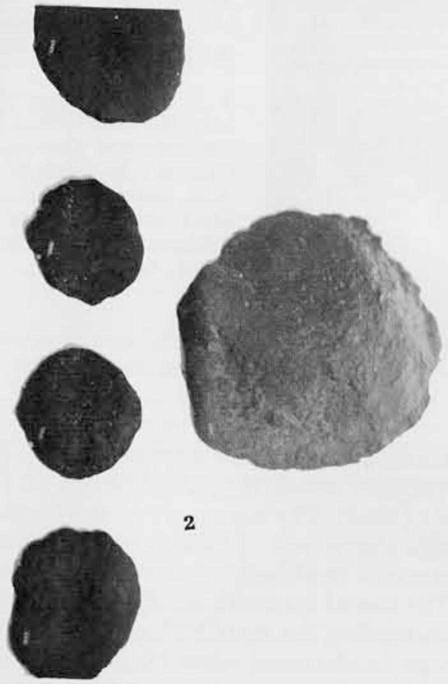
3
Oxhide ingots from Hagia Triada, as displayed in the Heraklion Museum. The Italian excavators found a total of 19 such ingots at Hagia Triada, all to be dated in the 16th century B.C.

4
Men from Keftiu (Crete) as shown in the tomb of the Egyptian official Rekhmire (Theban tomb no. 100) during the reign of the pharaoh Tuthmosis III (1490-1436 B.C.). Figure no. 1 bears an elephant tusk, no. 4 an oxhide ingot. Both materials were found together in a storeroom at Kato Zakro, on the east coast of Crete, excavated by Dr. Nicholas Platon. Ivory and copper were raw materials which the Minoans imported from the east. Drawing by Norman de Garis Davies.

5
Guard from Heraklion Museum carrying ingot from Hagia Triada. The ingot weighs about 30 kg., or over 60 lbs. which, in the system of weights used in the Bronze Age, represented one talent.

6, 7
General view and detail of one of the Hagia Triada ingots. The significance of the signs, which appear on a number of the oxhide ingots, has yet to be determined.

Photographs 2, 3, 5, 6, 7
by Ellen Herscher.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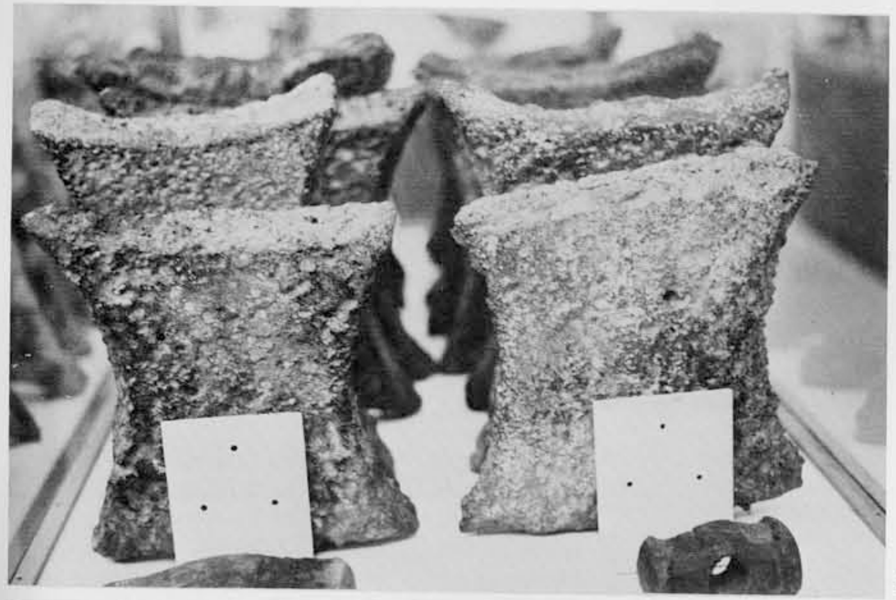
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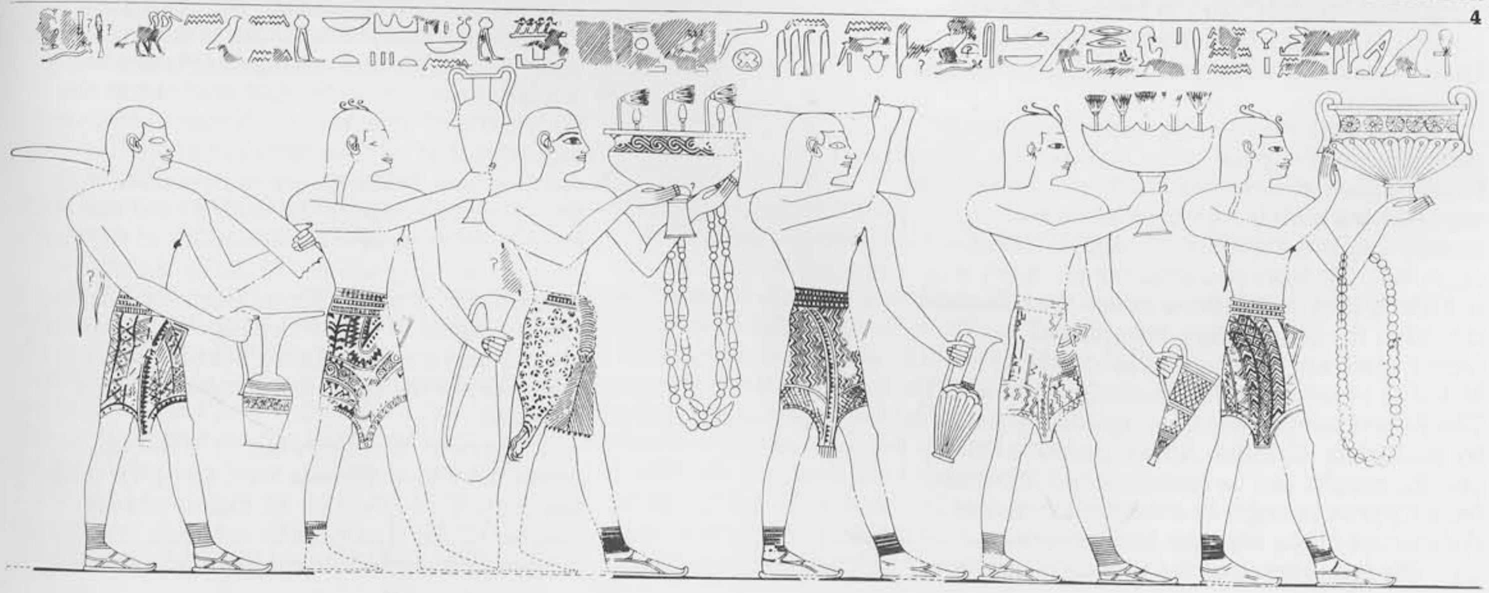
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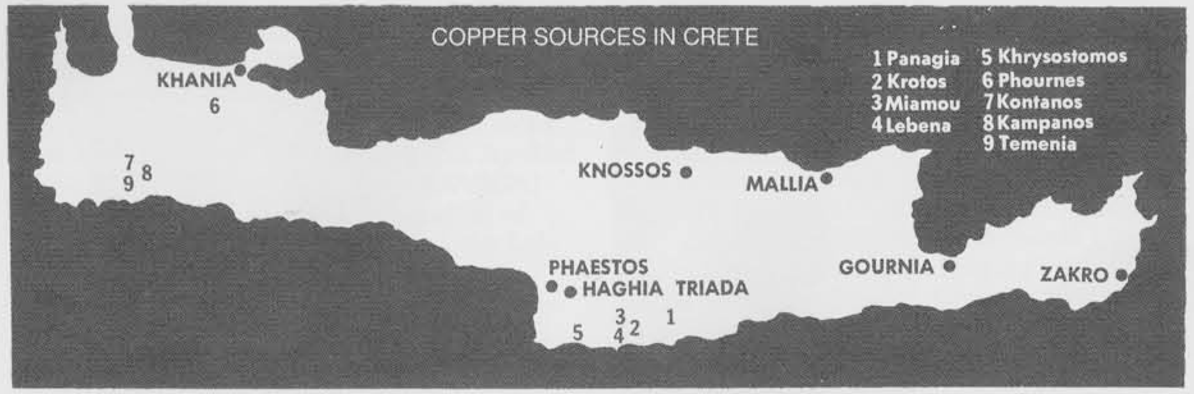

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1
Map showing possible copper deposits on Crete.

2
Map showing mining and metallurgical sites on Cyprus. After map in H. G. Buchholz, *Berliner Jahrbuch für Vor-und Frühgeschichte*, 7 (1967), p. 192, fig. 1a.

3
Map showing Mediterranean metallurgical sites.

2

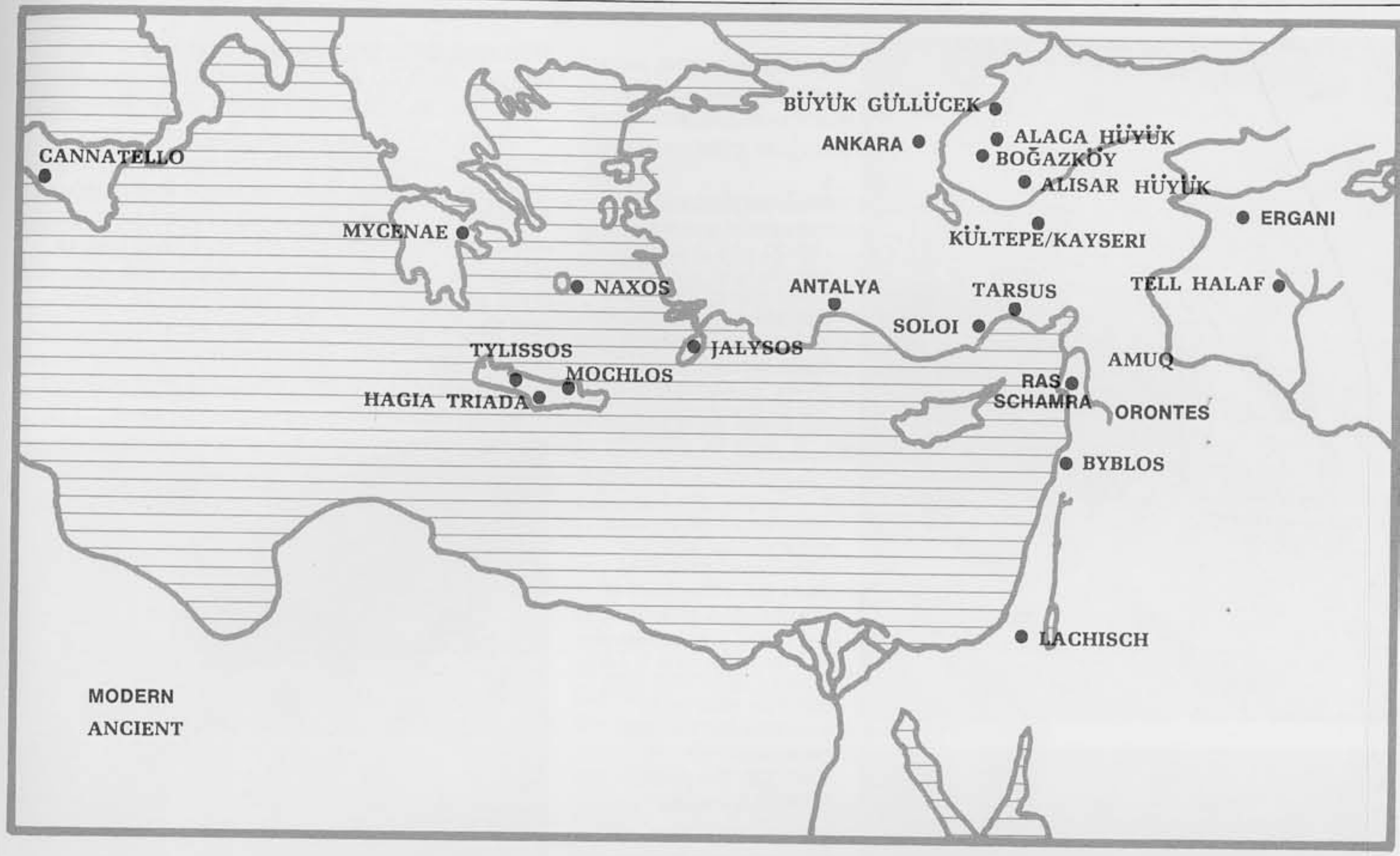
erately, rather than occurring in natural combination with copper. We do not at present have an index for the identification of Cypriote copper ores, although recent metallurgical (Steinberg and Koucky 1974) and geological studies (Constantinou and Govett 1973; Govett and Pantazis 1971) may assist in this endeavor. Cyprus cannot be discounted as one of the major sources of copper in antiquity; the distribution of Bronze Age archaeological sites along routes by which raw materials could reach the coast for shipment must be significant in terms of the economic life of the island (Catling 1969), and finds of slag and metal processing implements are explicit.

One must look also to Anatolia as a possible source of copper in the Bronze Age. The Ergani mines of southeastern Turkey probably supplied the metals for objects made at nearby Cayönü Tepesi in the eighth millennium B.C. (Çambel and Braidwood 1970); it is not likely that use of these mines was discontinued in the Bronze Age. Samples of wood from Ergani are now being carbon dated at MASCA to get some firm dates for the mines. The Ergani mines have been studied in detail by geologists (Griffitts, Albers and Öner 1972) and the results can be coordinated with those from Cyprus in order to determine whether differences in the ores can be discerned.

The importance of the oxhide ingots in reconstructing the ancient copper trade in the Mediterranean was first pointed out by H. G. Buchholz (1959). The discovery of the Cape Gelidonya shipwreck in 1960 dramatized the significance of Buchholz' observations (Bass 1967). The use of scientific analysis as an aid in understanding the distribution of these ingots was emphasized when Maddin and Muhly obtained a piece of one of the ingots from the Gelidonya shipwreck. Analysis of the ingot by emission spectroscopy revealed a significant cobalt content not present in any known Cypriote ores; a similar amount of cobalt has been detected in some ores from the Ergani mines of southeastern Turkey (Maddin and Muhly 1974). Since trace element composition may vary within a single ore body and not all Cypriote ores have been analyzed, this information is not sufficient evidence on which to base a revision of Bronze Age Mediterranean history, but it certainly indicates the need for analysis of more of the Gelidonya ingots and of Cypriote and Anatolian ores.

In September of 1974 drilled samples of seven ingots from four sites (Hagia Triada, Kato Zakro, Mochlos and Tylissos) were taken in the Heraklion Museum, through the kindness of Dr. Stylianos Alexiou, Miss Angelike Lembessi and Dr. Nicholas Platon. Preliminary analysis of these samples has been completed and arrangements are being made for neutron activation analysis at the University of Wisconsin. Elemental analysis indicates that all the ingots are of similar composition, being a very pure copper with no significant impurities. Both nickel and lead have been suggested as indicators of Cretan copper, but they are not found in the ingots studied. Since no cobalt was detected in the Cretan ingots, it might be suggested that the copper was derived from a source different from that of the Gelidonya ingot, perhaps Cyprus.

Three of the ingots found in the shipwreck off Kyme, Euboia were sampled with the support of Madame M. Oeconomides, curator of the Numismatic Museum in Athens. Although these ingots are heavily corroded



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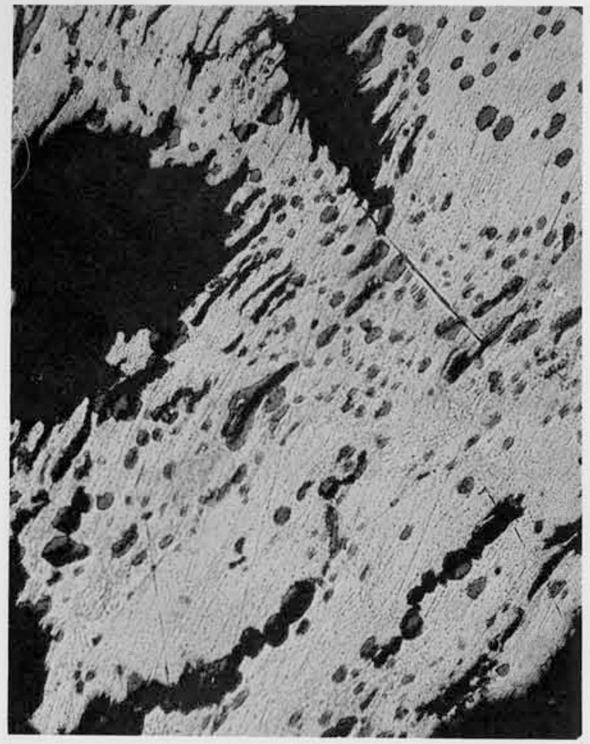
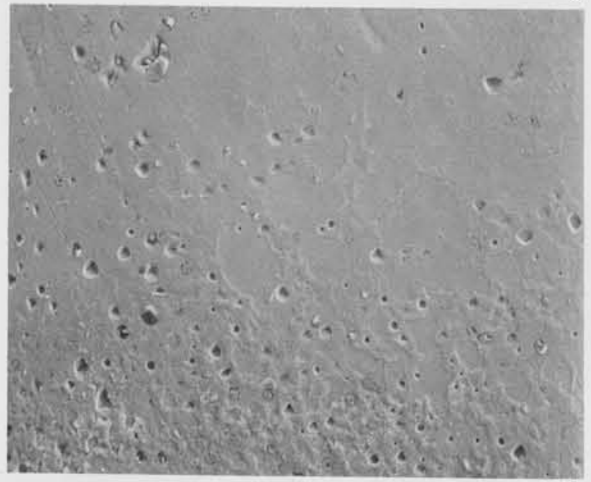
	Zakro	Hagia Triada	Hagia Triada	Hagia Triada	Mochlos	Mochlos	Tylissos	Kyme	Kyme	Kyme	Keos	Cape Gelidonya
Nickel (Ni)	.001	.003	.003	.005	.001	.005	.003	.005	.01	.005	.003	0.10
Cobalt (Co)	.003	.005	.10	.01	.003	.01	.003	.10	.01	.01	——	0.20
Bismuth (Bi)	.001	.001	.001	.001	.001	.001	.001	.001	.001	.001	——	——
Zinc (Zn)	.01	.01	.01	——	——	0.5	1.0	——	——	——	——	0.1
Aluminum (Al)	.03	.001	.001	.03	.001	.03	.03	.10	.10	.02	.10	0.01
Tin (Sn)	.001	.01	.001	——	.001	.001	.001	.003	——	——	——	0.05
Silver (Ag)	.001	.03	.001	.003	.001	.001	.001	.001	.001	.001	.05	0.08
Lead (Pb)	.001	.004	.003	.10	.001	.03	.10	.01	.01	.20	.010	0.07
Arsenic (As)	.013	.037	.10	.10	.03	.03	.03	.06	.02	.02	.001	0.23
Iron (Fe)	.05	.013	.014	.01	.001	.01	.01	0.5	0.5	.10	.28	10.
Magnesium (Mg)	.001	.001	.001	——	.001	——	——	——	——	——	.014	0.02
Silicon (Si)	.10	.10	.003	——	.003	——	——	——	——	——	.39	0.03
Manganese (Mn)	.001	.001	.001	.001	.001	.001	.001	.001	.001	.001	.025	0.005
Germanium (Ge)	——	——	.001	.001	.001	.001	.001	——	——	——	——	——
Molybdenum (Mo)	——	——	——	——	——	.001	.001	——	——	——	——	0.03
Sodium (Na)	——	——	——	——	——	.5	1.0	——	——	.03	——	0.1
Antimony (Sb)	——	——	.01	.01	.01	.01	.01	.01	.01	.01	——	0.02

with little remaining metal, elemental analysis did reveal a copper unlike that of the Gelidonya ingot. It was not possible to do any detailed metallographic examination since drilling had distorted the grain structure of the metal; there was nevertheless some sulphide detectable in the Kyme specimens. In contrast to the Kyme ingot, the ingot from the site of Ayia Irini on the island of Keos sampled through the kindness of Dr. John Caskey of the University of Cincinnati was essentially a pure copper. The Keos ingot is similar to the Cretan ingots in the purity of its copper and its metallographic structure.

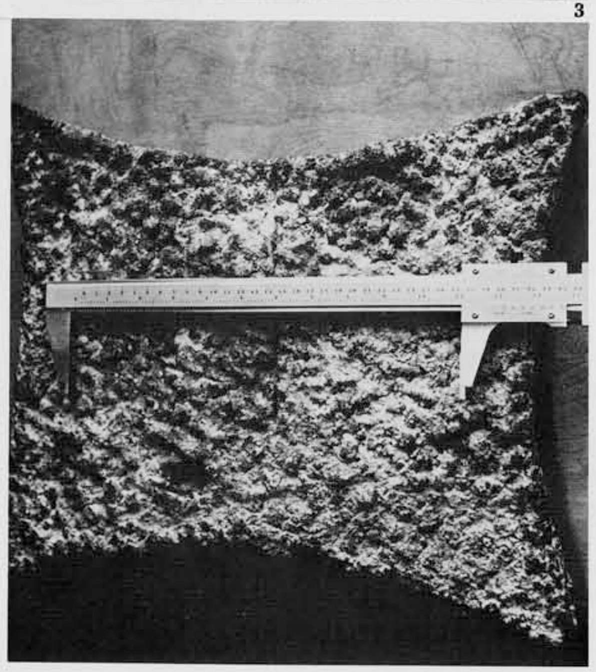
The only bronze oxhide ingot cited in archaeological literature is in the Metropolitan Museum of Art and may have come from a shipwreck in the Bay of Antalya, Turkey. An analysis performed in 1915 indicated that the ingot had a tin content of 7.5%, with some minor impurities (Richter 1915: No. 1810); this analysis is reported by Buchholz (1959:30). Through the kindness of the curator, Dr. Dietrich von Bothmer, and the assistant curator, Dr. Joan Mertens, we obtained samples of this ingot in January, 1975. Emis-



1
Photograph taken with the optical microscope (500 X) of a drilled sample from one of the Kyme ingots. The black patches are corrosion and the small grayish blobs are copper sulphide. The copper matrix shows the distortion caused by the drilling operation.



2
Micrograph taken with the scanning electron microscope (300 X) of a piece from fragment of oxhide ingot found at Ayia Irini, on the Island of Keos. The date is Late Minoan IB or 1500-1450 B.C. The even grain structure of this very pure copper shows clearly in this sample cut off with a hacksaw.



3
General view of oxhide ingot in The Metropolitan Museum of Art. This ingot was purchased from a dealer in 1911; it is of unknown date or provenance but, in size and shape, similar to the ingots found on Crete.

sion spectroscopy revealed that it is pure copper, with no tin and some minor impurities. Fine sulphide inclusions were detected with the use of the energy dispersive spectrometer associated with the scanning electron microscope. Thus the only ingot that might have been alloyed has been proved to consist of pure copper like the others. Other ingots allegedly from the Antalya shipwreck are cited as being in the Münzkabinett in Berlin and the Numismatic Museum in Athens (Parise 1968: 120); the latter is not in the museum at the present time and seems never to have been there.

4
Photograph taken with optical microscope (160 X) of sample taken from Metropolitan ingot. Again note the fine detail possible when working with an actual sample of metal. Here the grains of copper are outlined by copper sulphide which is also present within the grains themselves.

5
Micrograph taken with the scanning electron microscope (1400 X) of a drilled sample from one of the Hagia Triada ingots. The distorted grain structure is obvious, but it is possible to recognize particles of what must be copper sulphide.

Photographs 1, 2, 4, 5 and the three on the next page by Alexander Vaskelis.

A sample of one of the broken ingots found in the hoard of Mathiati on Cyprus was taken in the summer of 1973 through the courtesy of Dr. Vassos Karageorghis and Mr. K. Nicolaou. The sample was too corroded to be studied satisfactorily, but the copper seems similar to that of the Kyme ingots.

Although the results of the ingot study project indicate success in obtaining specimens for analysis, there are a number of problems which must be considered. To obtain samples, it is necessary to receive permission from governmental authorities; this must be done in person by direct negotiation. Ingots are not considered prime exhibition material by many museums and are often consigned to storerooms. It has been our experience that the ingots are sometimes misplaced and a considerable effort is needed to find them. Once they are located, the location and nature of the sample to be taken and methods of restoration must be discussed with museum curators and restorers. We have found that drillings are not good samples and that small cores must be taken. In the case of corroded objects, the corrosion layer may be gently scraped away in the area to be sampled and the core taken in the sound metal. Taking the cores requires a knowledge of metallurgical structures and properties in order to obtain the maximum sample with the least damage to the object.

Analysis of the specimens takes place at the Laboratory for Research on the Structure of Matter, with each sample treated according to an organized program of study. The specimens are prepared according to standard metallographic procedures of mounting, polishing and etching, and examined with the optical microscope. In the case of ingots, all of which were cast and were probably not subjected to any working, optical examination may indicate the environment in which the ingot cooled—a thin or thick mold or the bottom of a furnace. After complete optical study and photography, the specimens are examined by the scanning electron microscope. Particles in grain boundaries can be identified using the energy dispersive spectrometer associated with the scanner and the electron beam probe, and the results may indicate whether the ores used were weathered oxides or smelted sulphides and whether lead ores were added to improve the fluidity of the molten metal. Micrographs of the images generated by the scanner add to the documentation of each specimen. Optical emission spectroscopy determines the elemental composition of the sample. Neutron activation analysis, to "fingerprint" metals by determining trace elements in parts per million or billion, is important in the determination of ores used; similar amounts and consistent patterns of unusual trace elements in ores and ingots may indicate that the ingot metal derives from the ore source, although it must be borne in mind that trace element concentrations may vary within a single mineral deposit.

The analytical data provide the background for archaeological and historical conclusions about ingots and should help determine the role which ingots played in Bronze Age trade. The ingot was one form in which copper was shipped; scrap metal was also useful for fabrication of new objects. A typological survey of artifacts may indicate whether they were imported or manufactured in the area of their finding, although a much more extensive program of analysis of selected classes of material is necessary to confirm this.

Significant to the question of the ingots' role in the copper trade is a study of their archaeological contexts. Workshops and storerooms are the most logical locations in which to find ingots, so the shipwrecks of Kyme, Gelidonya and perhaps Antalya add an important dimension to the consideration of metallic raw materials. Another question to be considered relates to the availability of ingots. Were they disseminated through major ports, such as Kato Zakro, and thence distributed throughout the island of Crete? Did tinkers buy ingots or were they converted into smaller units at permanent metalworking sites? How were the ingots used? The example of the Israeli ingots indicates that larger masses of copper were sometimes cast into more convenient units before working of the metal took place. Further study of unfinished shapes may reveal other forms of secondary castings.

Two phases of the research project—Anatolia and the western Mediterranean—are crucial to the success of the ingot study. The significance of the Gelidonya ingots has already been discussed. The central Anatolian plano-convex ingots from Alaca Hüyük and Acemhüyük will provide an important counterpoint to the Gelidonya material. If any similarity of trace elements to the Gelidonya ingots exists, we might postulate two trade routes from the Ergani mines—one to the interior and the other to the coast.

The ingots found on Sicily and Sardinia are important because, as the westernmost representatives of the oxhide shape, their affinities need to be explored. They are usually studied as an adjunct to discussions of eastern Mediterranean ingots, certainly a crucial feature in their understanding (Parise 1968). Analytical work should allow us to place them in the proper context and perhaps to extend the range of the eastern Mediterranean trade routes. We will consider these ingots in the light of Italian and Iberian metal sources in an attempt to determine why ingots of this shape are found in the western Mediterranean.

Analysis of the ingots elucidates ancient mining practices. The Cretan ingots, as well as the one from Keos, date to the middle of the second millennium B.C. and show no significant pattern of sulphide inclusions. In contrast, the Gelidonya ingot of about 1200 B.C. contains massive copper-iron-sulphide particles. Earlier the copper ores used were weathered oxides, hydroxides and carbonates, such as azurite and malachite, which may contain only a small amount of sulphur. By the end of the Bronze Age the weathered surface deposits were sufficiently depleted so that miners were forced to go deeper into deposits where mixed ores containing more sulphide are found. Copper sulphide metallurgy requires a roasting process to oxidize the sulphur before smelting can take place; the presence of massive sulphide inclusions, many containing iron, and the porosity of the casting in some ingots probably indicate the imperfect nature of the earliest roasting operations.

Through the historical and scientific analysis of all ingots available for sampling in and from the Mediterranean area, we hope to develop an understanding of ancient trading patterns. Coordination of trace elements in the metal of the ingots, trace elements in Mediterranean ore sources and the locations in which ingots were found may enable us to follow the progress of ingots from mines and presumably nearby smelting sites to shipping depots to final destinations. If a number of ingots prove to have similar compositions, we can plot their distribution and suggest maritime and land routes along which the ingots were traded and possible trading mechanisms by which they were distributed. The definition of this one aspect of the ancient copper trade will provide the basis for further, more comprehensive programs of research into the early history of copper and bronze. 24

Tamara Stech Wheeler has a Ph.D. from the Department of Classical and Near Eastern Archaeology at Bryn Mawr College. Her special field is the archaeology of Bronze Age Anatolia and she has worked for a number of seasons with Dr. Machteld Mellink at the site of Karatas in southwestern Turkey. This past year she has been studying metallurgy in order to do analytical work on excavated objects of metal.

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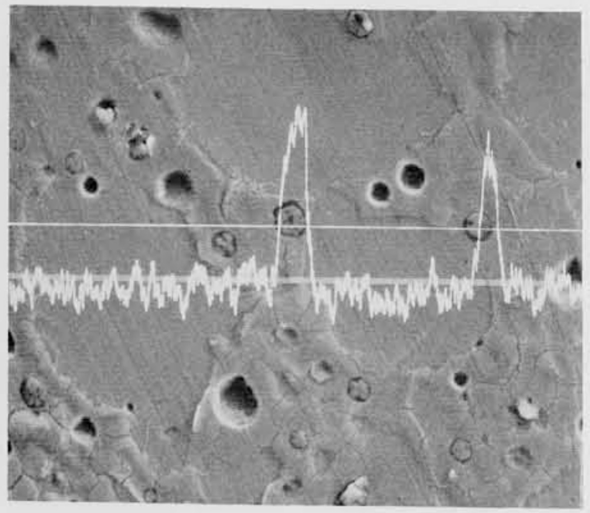
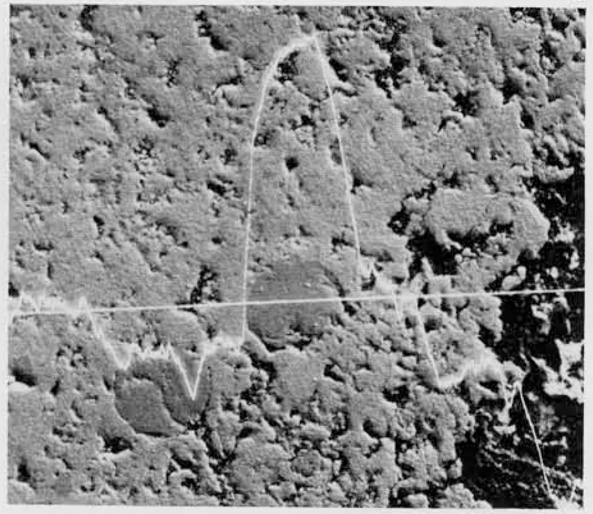


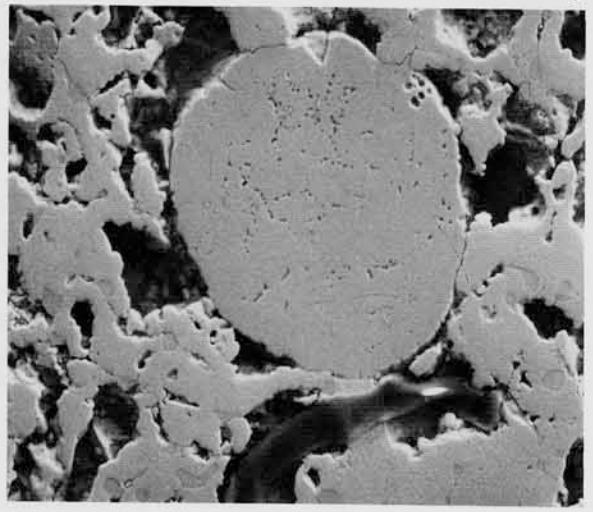
1
Sample from the Keos ingot studied with the scanning electron microscope (300 X), equipped with an energy dispursive spectrometer. This makes it possible to obtain qualitative chemical analysis of some of the "second phase" particles present in the copper. Here the spectrometer has been set to read for sulphur and the results indicate the presence of small particles of copper sulphide.

2



2
Same technique as in the previous illustration, here used to study a sample from one of the Cape Gelidonya ingots (100 X). The straight line represents the scan line and the wavy line corresponds to the energy of the sulphur present. The sharp rise in the sulphur line indicates a high concentration of sulphur, here present as large inclusions of copper-sulphide or *matte*. This *matte* is the result of the imperfect smelting of a mixed copper ore, containing both oxides and sulphides.

3



3
Detail of the massive inclusion of *matte* in the Cape Gelidonya ingot, as taken with the scanning electron microscope (300 X). This also shows the high porosity of the copper in the Cape Gelidonya ingot.

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James D. Muhly is Associate Professor of Ancient History at the University of Pennsylvania and the head of the Near East Division of the Oriental Studies Department. He has a Ph.D. from Yale University and formerly taught at the University of Minnesota. His research interests include the relations between Greece and the Near East in the Bronze Age and the importance of the metals trade in the development of Mediterranean commerce.

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