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METALLURGY, SOCIETY AND THE BRONZE/IRON TRANSITION IN THE EAST MEDITERRANEAN AND THE NEAR EAST

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INTRODUCTION

In so far as it refers to the chronological sequence of dominant technologies in the Old World, the three ages concept of archaeological classification cannot be challenged. Difficulties arise when attempts are made to specify precise boundaries between the epochs, both chronologically and technologically, and to associate distinctive cultural patterns with the technological phases. Some increase in cultural complexity was necessary for metallurgical innovation to be possible, but the material cultures and social orders were themselves drastically changed by the availability and use of metals, initially by an elite but later, in the Iron Age, by even the humblest ploughman in the fields. Furthermore, the fact that these changes took place at different times in different parts of the world impacts on the question of interactions and influences between ancient peoples, and on the argument of diffusion versus independent invention.

Waldbaum (1978) has surveyed the published archaeological finds of iron objects from the ancient Near East. She listed 14 items dated earlier than 3000 B.C., 8 between 2000 and 1600 B.C., and 74 corresponding to the Late Bronze Age, between 1600 and 1200 B.C. All the objects dated before 1600 B.C. were of jewellery or from prestigious contexts, pointing to the high value placed upon iron at that time. This contrasts with Sherratt's (1994) admittedly incomplete list of 199 iron objects from the 12th and 11th centuries in the Near East, not including 62 from Cypro-Geometric tombs at Kouklia-*Skales*.

A useful analysis of the long gestation time for the adoption of iron was put forward by Snodgrass (1971) who envisaged three stages in the progress of iron technology:-

Stage 1. Iron was used for ornaments and prestige items.

Stage 2. Iron began to be used for utilitarian objects but bronze still predominated.

Stage 3. Iron was in general use for utilitarian objects.

Towards the end of the 13th century B.C. the archaeological record of the Near East shows increasing use of iron for cutting tools and other working implements (McGovern 1987, Stech-Wheeler *et al.* 1981, McNutt 1990), gradually replacing bronze and even wood and stone for uses for which metal had hitherto been too costly a luxury, e.g. its use for ploughshares made possible the cultivation of ground previously considered too hard. In the Eastern Mediterranean this process occupied most of the 12th, 11th and 10th centuries, and is regarded as the transition from the Bronze Age to the Iron Age.

Early smelted iron was a rather unsatisfactory material, greatly inferior to bronze in most respects. It could not be melted and cast, but had to be consolidated by repeatedly heating and hammering to expel the slag and produce sinter fusion. The same process, known as forging, was used to fabricate objects. The metal thus produced, though harder than unalloyed copper, was much softer than bronze for cutting purposes, and rusted rapidly in a damp environment. Despite these failings, once a demand had been created for iron the abundance of its ores made it possible for the first time to produce a metal which was relatively cheap and widely available.

By the start of the 12th century the numbers of utilitarian objects made of iron (mostly in tombs) began to increase in Cyprus, Syria, Palestine and the Aegean (Làszló 1977, McNutt 1990, Sherratt 1994, Waldbaum 1978), though they were greatly outnumbered by those of bronze. During the Late Cypriot IIIA period (*c.* 1200-1170 B.C.) the trend towards iron intensified, and innovative types such as the iron pike from Tomb 417 at Lapithos showed adaptation to the new material. Carburisation and quenching, which have been identified in many of the single-edged iron knives produced in Cyprus at this time (Tholander 1971; Åström *et al.* 1986; Muhly *et al.* 1990) would have given a steel with a better cutting edge than bronze (Tholander 1971; Tylecote 1980).

Prior to the introduction of carburisation and quenching, and until the economic advantages of cheap, locally available raw materials were appreciated there would seem to be little reason for the widespread use of iron. These considerations have led to speculations that some external factors might have been responsible for the relatively rapid increase in iron production after such a long period of apparent indifference and for the social and economic changes which may have resulted. For some years the most popular idea was that propounded by Snodgrass (1971, 1980) and others, that a shortage of tin, caused either by exhaustion of known tin deposits or by disruption of international trade routes was the prime factor in promoting the use of this seemingly inferior substitute for bronze.

Other hypotheses have been postulated, such as fuel shortage (Wertime 1982), or Sherratt's sociological arguments (Sherratt 1994), and these will be discussed later. Meanwhile, the possibility of tin scarcity as a result of disruptions to mainly palace-based trading networks in the 13th-12th centuries is still open to evaluation through scientific analysis.

Waldbaum (1978) originally favoured the Snodgrass hypothesis, but suggested that it might be tested by chemical analyses of a substantial number of bronze artefacts dated to the period of the Bronze Age/Iron Age transition. A shortage of tin might be expected to show up as a reduced concentration of the element in the bronzes, with its possible total or partial replacement by other hardening additives such as arsenic, antimony, or zinc. The present work is a continuation of studies undertaken in response to Waldbaum's suggestion.

Initially a study was made of items from LC IIIA Kouklia in Western Cyprus (Pickles 1988). These analyses showed the tin contents to be generally high, with no suggestion of a shortage. However, they all related to one site in one part of the Near East, with no known association with tin production. It was clearly desirable to extend the study to sites in other regions to see if a consistent pattern were maintained.

Inevitably, the availability of archaeological material dictated the scope for further work, and the authors thank Prof. Hartmut Kühne for making available the whole of the bronze finds from his excavations at the Syrian sites of Tell Bderi and Tell Sheikh Hamad, Prof. George Bass for material from the Cape Gelidonya shipwreck, Prof. Paul Åström for the Hala Sultan Tekke (Cyprus) bronzes and Dr Jonathan Tubb for access to the bronzes from the British Museum excavations at Tell es Sa'idiyeh (Fig. 1).

A catalogue of the items analysed is given in Table 1.

THE SITES

Tell Bderi and Tell Sheikh Hamad

Rescue excavations at Tell Sheikh Hamad and Tell Bderi, in eastern Syria, were part of an interdisciplinary research project under the direction of Prof. Dr Hartmut Kühne of the Freie Universität Berlin, prior to the construction of the Habur hydroelectric reservoir (Kühne 1983, 1984, 1986/7, 1989/90, 1990, 1991).

Tell Bderi is located *c.*25km. south of the provincial capital of Hasaka, on the right bank of the Habur river. The excavation was undertaken by Prof. Dr Peter Pfälzner (1989/90). In the first half of the Early Bronze Age (Early Dynastic I/II) the site held a large walled town which, between Early Dynastic II/III to the start of Akkadian times, spread to an area of 5-6 hectares, covered mainly by interconnecting, small-roomed houses. The latest 3rd millennium houses were all destroyed at the same time, after which a hiatus in the Middle Bronze Age was followed by a revival in the Late Bronze Age, with more complex, larger-roomed houses than in the EBA town.

In the Middle Assyrian period a deep, rectilinear pit 4.5×6m. was sunk to the west of a Mittanian road. At a depth of 11 meters the bottom had still not been reached. The fill was uniform from top to bottom, and fragments of a foundation cylinder show that the pit was all filled at the same time. Builders' marks give the town name Dùr-Assur-kitte-lisir and inscribed burned clay cylinders provide a *terminus post quem* for the fill as the reign of Tiglath-Pileser I (1114-1076 B.C.).

Table 1. Catalogue of analysed bronze objects.

Cat. No.	ITEM	CONTEXT	DESCRIPTION	PERIOD
			OBJECTS FROM TELL BDERI	
1	BD 149	2745 II ;T2,	Pendant, fairly large	1114 - 1076 BC
2	BD 346	2745 II ; FS314	Fibula (?), Middle Assyrian	1114 - 1076 BC
3	BD 198	2745 II ; FS277	Fragment (rod?)	1114 - 1076 BC
4	BD 224	2745 II ; T2, FS247	Fragment of sheet 15 x 3 mm	1114 - 1076 BC
5	BD 275	2745 ; T2, FS247	Pin or nail (2 fragments)	1114 - 1076 BC
6	BD 252	2745 III ; T3, FS24	Pin or nail (fragment)	1114 - 1076 BC
			OBJECTS FROM TELL SHEIKH HAMAD	
7	SH ? 100	1527 IV ; Grave 9,FS5	Fibula fragment	C9 BC
8	SH83 139	8977 IV ; T10, M45	Massive fragment.	C7 BC
9	SH83 86	9145 III ; T10, M45	Massive fragment.	C7 BC
10	SH83 24	9177 IV ;Corner,Rm B	Fragment 20x20x7 mm.	C7 BC
11	SH87 156	9177 ; Rm E, FS116	Hook	C7 BC
12	SH82 32	8977 II ; T5, G8	Thick unidentified fragment	C7 BC
	Items 13 to 17 are of later date and not relevant to the main theme of the paper, but are included for completeness			
13	SH78 21	9791 III ; Btw. S & SE cols	Fragment	c. 100 AD
14	SH80 9	1927 IV ; T2 section	Part of fibula with iron pin.	Roman
15	SH87 51	5751 I ; FS75	Bracelet, dia. 60 mm.	Roman
16	SH80 88	1927 I ; T6, deepening	Pincer	C2/3 AD
17	SH78 4	1729 III ;Canal 1 section	Heavy pin, 3 pieces	?
			OBJECTS FROM CAPE GELIDONYA	
18	24	BWG 50	Cylindrical piece (Fragment.)	*c*. 1200 BC
19	26	CG '89	Fragment	*c*. 1200 BC
20	417	BWG	Hoe fragment	*c*. 1200 BC
21	37	CG A or 9	Possible adze fragment.	*c*. 1200 BC
			OBJECTS FROM HALA SULTAN TEKKE	
22	N1018 (III)	A.8, layer 4	Chisel fragment, 1.7x 0.5 x 0.4 mm.	LC IIIA
23	N6004 (V)	A22,Layer 1	Chisel 7x 0.5 x 0.5 mm.	LX IIIA
24	N2040 (VIII)	A6s,Layer 2	Tweezers (?) 2.3 x 0.8 x 0.6 mm.	LC IIIA - LC IIIA-2
25	1981 F1602	W bottom L3	Bronze strip	LC IIIA
26	1979 F1358	Area 8	Fragment	LC IIIA
27	N 787	Well 1750	Pin or nail (?)	LC IIIA
			OTHER OBJECTS FROM CYPRUS	
28	L6	Unprov. Liverpool Museum.	Tripod	c.1200 BC
29	L10	Kouklia Marchello	Cauldron, from siege tunnel	Cypro-Archaic - (497 BC)
			OBJECTS FROM TELL es-SA'IDIYEH	
30	T51 - 1	BB600	Anklet	c. 1200 BC
31	T51 - 3	BB600	Anklet	c. 1200 BC
32	T51 - 4	BB600	Anklet	c. 1200 BC
33	T56 - 1	BB100	Bracelet	c. 1200 BC
34	T176 - 2	BB200	Bracelet	c. 1200 BC
35	T176 - 3	BB200	Bracelet	c. 1200 BC
36	T176 - 4	BB200	Bracelet	c. 1200 BC
37	T46 - 10	BB200	Bowl	c. 1200 BC
38	T222 - 1	BB600	Bowl	c. 1200 BC
39	T46 - 11	BB200	Dagger	c. 1200 BC
40	T204 - 8	BB100	Knife	c. 1200 BC

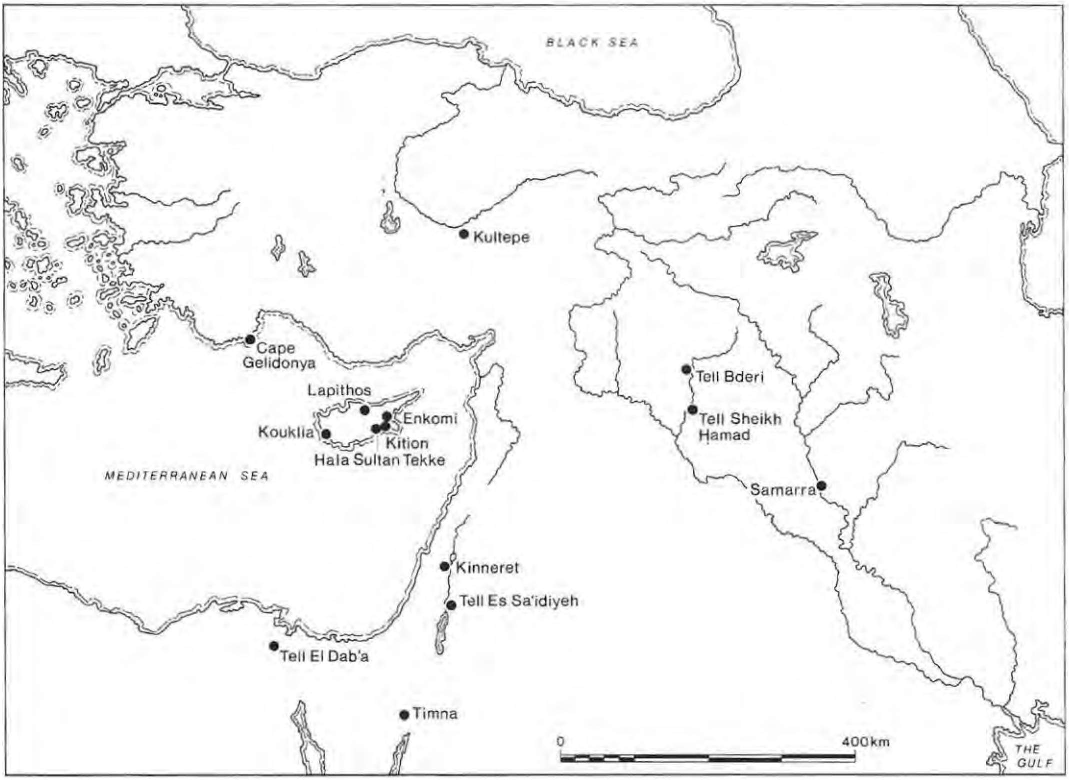

Figure 1. Map of East Mediterranean and Near East showing location of places mentioned in the text.

From this pit eleven metal fragments were recovered, of which six were analysed, despite a significant degree of corrosion.

Tell Sheikh Hamad is the site of the Neo-Assyrian town of Dùr-katlimmu (Kühne 1983), though evidence of occupation extends from the Late Bronze Age to Roman times. It is situated on the bank of the Habur river, some 120 km south of Hasaka.

The excavation seasons of 1978 through 1983 yielded 119 metal objects, of which 45 were unstratified stray finds, including 35 coins. Only 11 of the remaining 74 items were judged suitable for analysis, the others being too small and fragile, or too corroded. The selection was based purely on the physical condition of the objects, as full dating information was not known at the time. When this was forthcoming the dates were seen to range from the 9th century B.C. to the 2nd/3rd century A.D. The analyses of the later items (**Cat. 13-17** inclusive) are not relevant to the main theme of this paper, but they are reported here for completeness.

Cape Gelidonya

The southern coast of Turkey has through the ages been the scene of many shipwrecks, and numerous archaeological treasures have been recovered by the local sponge fishermen. One such, a bronze bust of Demeter, was seen by P. Throckmorton in 1953, and led to the recording of over 30 wrecks. At Anadolu Burnu or Cape Gelidonya, a bronze spear and a bronze knife suggested a shipwreck of pre-Roman date, and the first of a number of seasons of excavation was begun by G. F. Bass and P. Throckmorton in June 1960.

The main report by Bass (1967) reveals that the timber of the ship had almost completely disappeared, but a length of about 10 metres could be deduced from the position of the cargo. This consisted mainly of copper and bronze ingots and bronze scrap. There had also been some tin ingots which had been preserved as heaps of tin oxide only where covered by bronze or copper and concretion.

The ship was deduced by Bass to have been of Syrian origin on the basis of the artefacts of a personal nature ascribed to the captain's quarters. On stylistic grounds most of the metal objects had Cypriot parallels, though some of the hoes, adzes and the mattock had affinities with examples from Anatolia. The pottery, thought to have been mainly for the crew's use, was typical of a wide geographical area within the period LC IIB to LC IIIA (1350-1150 B.C.). Carbon dating of twigs used for packing the cargo gave a date of 1200±50 B.C. after dendrochronological calibration.

Hala Sultan Tekke

Situated near the Larnaca Salt Lake in south-east Cyprus, Hala Sultan Tekke is believed to have been an important commercial centre in the Late Bronze Age. The first excavations by the British Museum were never published in detail but they are well summarised in the eight volume report by Paul Åström *et al.* (1976-83), which mainly covers the more recent excavations under Åström's direction.

Bronze working seems to have been a significant activity, and the composition of slag reported by Hans-Gert Bachmann (1974,1976) indicates that smelting took place. The high iron oxide to silicate ratio suggests either addition of too much haematite flux or failure to add sand (silica) to an iron-rich copper ore. In either case there would be a potential for the accidental formation of iron if the smelting conditions varied.

Of the objects analysed here, three came from the Larnaca Museum store and are published in the Hala Sultan Tekke report, while three more or less corroded items came from the Dig House store near the excavation site. At least five of the objects could be classified as tools or implements, while the sixth (**Cat. 26** in Tables 1, 2 and 3) was unidentified. All were dated to the period LC IIIA to LC IIIA-2.

Tell es-Sa'idiyeh

This is a large double mound with a summit area of about 10 hectares, on the south side of the Wadi Kufrinjeh in the Central Jordan Valley, 1.8 km east of the Jordan river. The main occupation evidence covers the period from EB I-II to the Persian period, with some extensions into Roman and Byzantine periods, and traces of nearby Chalcolithic occupation.

The first excavations by the University of Pennsylvania in 1965-1967 (Pritchard 1980, 1985) revealed on the upper mound strata of the 10th to 8th centuries B.C. On the lower mound immediately below the surface, was found a large cemetery of the LBA to Iron I transition period (13th to 12th centuries B.C.), which cut into an Early Bronze Age occupation layer.

Excavations were resumed twenty years later, directed by Dr Jonathan Tubb for the British Museum (Tubb 1988a, 1990; Tubb and Dorrell 1991, 1993, 1994). The work revealed a complex walled city of the 13th to 12th centuries on the upper tell, with which the cemetery on the lower tell was associated. Both areas showed some evidence to indicate that the site was under direct Egyptian control during this period. The walled city appears to have been destroyed by fire around 1150 B.C.

The bronze items whose analyses are reported here came from the cemetery on the lower tell. Many of the graves were simple pit burials but others were better made, with mud-brick linings and grave goods.

The grave inventories for the British Museum excavations up to 1993 list 186 metal objects of which 139 were of bronze, 31 of iron, 10 silver, 5 gold and one composite iron and bronze. As the chronology of the graves coincided roughly with that accepted for the start of the Iron Age in the East Mediterranean, the inventory was sorted into groups according to the presence or absence of iron objects in the graves, to separate those which might possibly be Bronze Age from those which were unambiguously Iron Age. Thus, Group A were from graves with bronze but no iron objects, Group B from graves with bronze objects and iron ornaments or jewellery, and Group C from graves with bronze objects and iron tools or weapons. It was hoped that a selection of bronzes from each group and artefact type would have a good chance of spanning the Bronze Age/Iron Age transition and exposing any changes in bronze-making practices.

Unfortunately, despite the seemingly large number of items recovered in the excavations there were very few in Groups B and C, and overall the number available for analysis was drastically reduced by a combination of factors. A few of the graves in the Inventories were of Persian/Iron III date; very many of the LBA/Iron I artefacts were held in Amman, and some were on public exhibition in the British Museum. Of the remainder, many were too fragile or too small or too corroded to permit microdrilling for analytical samples. In the event only eleven samples were available for analysis, of which five had slight corrosion and two significant corrosion. None of the samples were from Groups B or C.

Since these analyses were carried out, continued excavation at Sa'idiyeh (Tubb *et al.* 1996 and 1997) has revealed more graves of the Late Bronze/Early Iron Age containing metal artefacts which would surely repay further study.

THE ANALYSES

There are various methods available for determining the compositions of ancient bronze. Traditional chemical analysis was considered too demanding of time, labour and materials, so an instrumental method was sought. Non-destructive methods such as X-ray fluorescence have the disadvantage of very low precision and measure only the surface layer, which is rarely representative of the bulk composition. Other techniques require the removal of some metal from the artefact, which is usually achieved by micro-drilling, using a 1.5mm. carbon steel bit to obtain a sample mass of about 10 to 30 milligrams. The initial drillings are discarded as being possibly affected by surface corrosion.

In the work previously reported the samples thus obtained were analysed by the neutron activation technique, in which many of the constituent elements, when exposed to the high neutron flux of a nuclear reactor, are converted into radioactive daughter products whose characteristic radiations can be measured. Although excellent for trace elements in ceramics, the method is less satisfactory for bronzes where the major component copper produces a very high output of penetrating, short-lived radiation, swamping that from other short-lived isotopes and presenting a handling hazard to the operator.

Spectrographic methods are probably the most satisfactory when the sample size is limited.. These depend on the optical properties of elements in the vapour phase, whereby, depending on the degree of excitation, they absorb or emit light of characteristic wavelengths. In the absorption method (AAS or atomic absorption spectrography) monochromatic light of the chosen wavelength is passed through a hot zone (flame or graphite furnace) into which a solution of the sample is sprayed. The reduction in intensity of the emergent light gives a measure of the concentration of the element with the corresponding absorption band. A high accuracy is possible, but only one element at a time can be measured. Emission spectrography has the advantage of allowing the measurement of a large number of elements in a single scan, but it requires a higher degree of atomic excitation which is usually achieved by raising the sample to a much higher temperature, as in Arc or Inductively Coupled Plasma (IPC) Spectrography. The high temperatures also produce substantial ionization, which causes a high background noise and reduced sensitivity. A new technique, Furnace Atomic Non-thermal Excitation Spectroscopy or FANES, has been devised (Falk, Hoffmann and Ludke 1983) to overcome this disadvantage by using a high voltage electrical discharge to produce the excitation needed for emission spectroscopy at the temperature of the AAS graphite furnace.

For the present work it had been hoped to use FANES or ICPS, but in both cases the equipment available was in an experimental stage and proved to be unsatisfactory. The analyses were therefore carried out using a commercial AAS instrument, which gave excellent results exceeding all expectations in terms of precision, sensitivity, and economy of sample consumption

The method used was basically that described by Hughes, Cowell and Craddock (1976) except that aqua regia was used instead of nitric acid for the dissolution of the samples. All the bronze samples dissolved completely, though on dilution of the concentrated solutions of some of the severely corroded material a slight cloudiness formed which, in the worst cases, became a white precipitate on standing. This was filtered off and identified as metastannic acid. Since no such cloudiness was produced from un-corroded bronzes even where the tin content was very high, it is concluded that the acid-soluble alpha-metastannic is formed by the action of aqua regia on the corrosion products of tin but not on tin metal. Dilution presumably changes the metastannic acid to the more stable and very insoluble beta-form.

A further variation on the British Museum method applied to the gold determinations, for which a palladium/hydroxylamine matrix modifier was used in accordance with the method of Hall and Vaive (1989)

As a check on possible experimental and operator variables an inter-laboratory comparison was arranged with the British Museum Research Laboratories, who provided standard bronzes for analysis. The results showed excellent agreement as shown in Table 6. Routinely, one of the standard bronzes was then included with every group of archaeological bronzes analysed.

The results are considered in Table 2, site by site, and according to artefact type.

SITE BY SITE SURVEY OF THE RESULTS

Tell Bderi

The rather severe corrosion of many of the objects causes uncertainty about their exact com-

Table 2. Analytical results

CAT.					ELEMENT %									
NO.	**Cu**	**Sn**	**As**	**Sb**	**Pb**	**Zn**	**Fe**	**Ag**	**Au**	**Ni**	**Co**	**Mn**	**Cd**	**Bi**	**Total**
BRONZES FROM TELL BDERI															
1	65.19	0.76	0.447	0.042	14.16	14.59	0.088	0.181	0.0041	0.046	0.008	0.002	0.001	0.001	95.5
2	84.99	2.11	1.047	0.078	0.265	0.018	0.313	0.040	0.003	0.155	0.006	0.001	0	0	89.0
3	60.04	3.53	0.287	0.009	0.035	0.020	0.258	0.0004	0	0.003	0.010	0.004	0	0	64.2
4	61.37	4.40	0.104	0.039	0.062	0.002	0.289	0.006	0.0001	0.008	0.005	0.004	0	0	66.3
5	53.42	0	0.506	0.036	0.056	0.011	0.640	0.0014	0.0004	0.077	0.006	0.002	0.200	0	55.0
6	74.32	1.55	0	0.012	0.006	0.003	0.109	0.0002	0.0	0.022	0.012	0	0.0008	0.01	76.0
BRONZES FROM TELL SHEIKH HAMAD															
7	67.00	3.11	0	0.012	0.103	0.327	0.123	0.033	0.0008	0.018	0.003	0	0.0024	0	70.7
8	84.65	0.15	0.711	0.716	3.67	2.71	1.17	0.129	0.008	0.017	0.005	0.003	0.002	0.022	96.0
9	88.89	0.21	0.24	0.04	0.008	0.031	0.044	0	0.0005	0.672	0.009	0.001	0	0.0023	90.1
10	66.08	0.06	2.99	2.36	12.86	2.47	0.714	0.217	0.0017	0.013	0.003	0.001	0.0087	0.091	87.9
11	85.59	1.10	0.14	0.04	8.22	0.006	0.030	0.033	0.010	0.063	0.021	0	0.0003	0.0024	95.3
12	86.17	0.05	3.33	0.213	1.23	1.28	2.32	0.137	0.0067	0.030	0.012	0	0.0012	0.0610	94.8
	The following items 13 to 17 are of later date and are not relevant to the main theme of the paper but are included for completeness														
13	65.12	4.52	0.018	0.322	0.488	0.018	0.014	0.308	0.0016	0.033	0.012	0	0.0056	0.018	70.9
14	63.66	2.51	0	0.054	22.87	0.227	0.103	0.253	0.0022	0.046	0.008	0	0.0016	0.0074	89.7
15	82.08	9.69	0	0.072	1.10	0.012	0.246	0.067	0.0012	0.053	0.006	0.001	0.0054	0.035	93.4
16	48.15	0.35	0	0.018	0.169	2.63	0.571	0.021	0	0.180	0.003	0.008	0.0025	0.0012	52.1
17	58.58	3.38	0	0.004	0.172	0.013	0.100	0.011	0.0012	0.013	0.014	0	0.022	0	62.3
BRONZES FROM CAPE GELIDONYA															
18	89.06	6.89	0.741	0.001	0..074	0.007	0.406	0.011	0.0009	0.027	0.0734	0.002	0.0013	0.0007	97.3
19	84.41	8.12	0.746	0.004	0.031	0.010	0.348	0.009	0.0011	0.050	0.067	0.0033	0.0013	0	93.8
20	86.14	8.07	0.639	0.008	0.022	0.007	0.177	0.009	0.001	0.057	0.074	0.002	0.0019	0	95.2
21	69.27	6.88	0.456	0.004	0.078	0.002	0.110	0.004	0.001	0.045	0.048	0.001	0.0146	0	76.9
BRONZES FROM HALA SULTAN TEKKE															
22	91.33	7.99	0.422	0.040	0.031	0.005	0.307	0.0063	0.0012	0.022	0.011	0.002	0.0115	0	100.2
23	92.02	5.27	0.936	0.045	0.100	0.005	0.228	0.0060	0.0005	0.032	0.006	0	0.006	0	98.7
24	91.66	4.99	0.736	0.044	0.016	0.009	0.240	0.011	0.0016	0.050	0.218	0	0.0006	0	98.0
25	52.40	3.92	0.050	0.021	0.060	0	0.257	0.003	0.0011	0.020	0.009	0.007	0	0	56.7
26	67.85	5.33	0.265	0.002	0.540	0.005	0.438	0.009	0.0016	0.019	0.018	0.002	0.0012	0	74.5
27	82.55	0.49	0.238	0.016	0.001	0	0.629	0.005	0.0009	0.047	0.099	0.005	0.0002	0	84.1
OTHER BRONZES FROM CYPRUS															
28	93.50	1.89	0.397	0.009	3.77	0	0.024	0.007	0.0015	0.031	0.002	0	0	0	99.6
29	98.4	0.08	0.679	0.005	0.014	0	0.027	0.0389	0.0082	0.011	-	0	-	0	99.3
BRONZES FROM TELL es-SA'IDIYEH															
30	87.7	9.11	0.17	0.0	0.83	0	0.041	0.014	0.0416	0.048	-	0	-	0	98.0
31	82.9	12.2	0.012	0.0	1.11	0	0.030	0.006	0.0026	0.038	-	0	-	0	96.3
32	79.7	10.9	0.06	0.0	0.92	0	0.057	0.003	0.0007	0.053	-	0	-	0	91.7
33	80.3	13.3	0.16	0.0	1.52	0	0.051	0.012	0.0017	0.033	-	0	-	0	95.4
34	85.6	10.5	0.12	0.036	0.88	0	0.064	0.003	0.0015	0.033	-	0	-	0	97.2
35	87.8	10.3	0.070	0.0	0.56	0	0.081	0.005	0.0011	0.037	-	0	-	0	98.9
36	87.6	9.62	0.097	0.009	1.20	0	0.085	0.010	0.0023	0.046	-	0	-	0	98.7
37	63.6	16.3	0.54	0.015	0.19	0	0..061	0.024	0.0026	0.053	-	0	-	0	80.8
38	77.5	13.8	0.35	0.009	0.095	0	0.068	0.011	0.0021	0.047	-	0	-	0	91.9
39	77.6	8.13	0.41	0.011	0.43	0	0.062	0.007	0.0038	0.053		0	-	0	86.7
40	86.5	8.68	0.36	0.030	0.13	0	0.200	0.005	0.0021	0.043	-	0	-	0	96.0

positions, but all contained at least one alloying element at a concentration greater than would be expected for natural impurity, although well below the level needed for full hardening. In the light of Moorey's observation (1985) that most metal tools and weapons in 2nd millennium B.C. Mesopotamia are of unalloyed copper (but see Shell 1997), these analyses cannot be cited as indicating an unusual shortage of tin. Four of the objects contained both arsenic and tin, albeit in small amounts, which might hint at the recycling of scrap.

Cat. 1, a pendant, was almost free from corrosion, and the analysis therefore accurately represents the original composition. It had a very strange composition, difficult to explain on metallurgical grounds, with over 14% each of lead and zinc. Until the discovery of the cementation process at the end of the second millennium B.C., zinc was produced only with great difficulty and probably accidentally, perhaps in the flues of silver/lead smelting furnaces (Craddock 1988) and was a costly material, rare as a major additive before the first century B.C. (Craddock and Gumlia-Mair 1988). However, the possibility that the pendant might be of a much later date than the other items is rejected by the excavator (H. Kühne, personal communication) because of the very secure context in which it was found. The coincidence of such high levels of zinc and lead (which must have been added in metallic form) arouses speculation that the zinc might have been produced in the flue of the lead smelter.

Tell Sheikh Hamad

The objects from this site cover a wide chronological span from the 9th century B.C. to the 2nd century A.D., and the pattern of analyses points to continued poor supplies of tin in the first half of the first millennium B.C. The ninth century fibula was a low tin bronze comparable with some of the Tell Bderi material, but tin was almost absent in the 7th century items. Of these, **Cat. 9** was basically unalloyed copper while arsenic and antimony (twice in almost equal proportions) would have given various degrees of hardening to **Cat. 8, 10** and **12**, which also contain significant amounts of zinc. Lead was also present in what would, in two cases, be considered unnecessarily high concentrations (12.9 and 8.2%), but this was exceeded in the Roman pendant which had an astonishing 22.9% lead. However, very large additions of lead, up to 20-30%, were sometimes used for intricate castings to take advantage of the extended plastic range and lower melting point (Northover 1988). Lead was also thought to be added for cheapness where strength was not important.

From the end of the first millennium B.C., tin bronzes seem to have been in use at the site, but only the Roman bracelet (**Cat. 15**) could be regarded as a high tin bronze.

Overall, the analyses endorse Moorey's (1985) observation of little tin usage in Northern Mesopotamia throughout much of the last two millennia B.C., but the erratic compositions might suggest attempts to develop a satisfactory alloying technology.

Cape Gelidonya

These bronzes show remarkably uniform compositions, especially since they are believed to form part of a load of scrap metal for recycling. The tin contents are between 7 and 8%, i.e. at the lower end of the optimum range. In addition they all contain around 0.6% arsenic, which is probably insufficient to confirm deliberate addition, and would have only slight hardening effect on its own. Other trace impurities are insignificant.

The suggestion is that these four items came from the same source and were made from new, i.e. unrecyled bronze, and it will be interesting to see if this uniformity is maintained when more analyses of the collection become available.

Hala Sultan Tekke

Cat. 22 to **26** proved to be tin bronzes with (allowing for the corrosion of **Cat. 25** and **26**) between 5 and 8 % tin —at the lower end of the optimum range for full hardening. Arsenic was also present, mostly at a level above average for natural impurity in the ore but below that needed for full hardening on its own. Its presence might indicate either an attempt to economise in tin usage or the recycling of arsenical bronze. By the Late Bronze Age it might be supposed that the deadly effect of arsenic on the metalworkers would be known, so the second explanation would be more reasonable. Levels of minor components were low and fairly uniform. The remaining, unidentified, object (**Cat. 26**) which came from a LC IIIA context in Well 1750, had only about 0.5% tin and 0.25% arsenic, which would not give significant hardening.

The relatively small spread of compositions seems to indicate well-controlled operation of the bronze production. Tin was clearly available but its use was not profligate, which could indicate either sound economics or some constraints on tin supplies.

It is interesting to note that the use of tin was more extravagant in the items from Kouklia than in these from Hala Sultan Tekke (cf. Pickles 1988). This might reflect the different contexts (i.e. graves vs. occupation areas) or signify local industries with independent contacts.

Tell es-Sa'idiyeh

There was clearly no shortage of tin, all the analyses being above the recommended minimum of 7.0%, and mostly around the 10-12% of modern practice. Correspondingly there was no apparent attempt to replace tin as a hardener by other metals such as arsenic, antimony or zinc. However, the last four artefacts in the table contained low levels of arsenic, barely high enough to suggest the possibility of deliberate addition, while the group of anklets and bracelets all contained an average of 1% of lead (which is sometimes added-though ideally in about twice that amount (Craddock and Gumlia-Mair 1988) —to facilitate casting) and no arsenic. This might indicate the different preferences of different bronze founders.

Trace impurities were generally low and uniform, the low levels of iron suggesting efficient and well-controlled smelting techniques. Possibly significant traces of gold in the anklet 51-1 (416 parts per million) and of silver in the bowl 46-10 (240 parts per million) might have derived from surface decoration. Their single and separate occurrences make it unlikely that they came from natural impurities in the copper or tin ores.

Although there must be uncertainty about the exact tin: copper ratios in the two highly corroded objects (which both came from the same grave), there was again no deficit of tin, and the minor elements conform closely with the general pattern of these analyses. The metastannic acid precipitate was almost certainly produced by the corrosion of the manufactured artefacts, and dissolved initially because of the use of *aqua regia* instead of nitric acid for the dissolution.

The most striking characteristic of the analyses is the relative homogeneity of the compositions, with no indications of the recycling of scrap bronze. The excavation reports cited do not reveal evidence of bronze manufacture at Tell es-Sa'idiyeh, although on such a large site, at present only partially excavated, the possibility cannot be excluded. Whether or not the bronzes were locally manufactured, the homogeneity of the compositions indicates a consistent source of raw materials and a well-controlled technology. As tin is variable in Egyptian bronzes, this consistency bears on the nature of Egyptian control of copper sources and metalworking in general in this area.

CLASSIFICATION OF RESULTS ACCORDING TO ARTEFACT TYPE

Twenty-nine of the analyses reported in Table 2 were of identifiable objects, and these have been abstracted, grouped according to artefact type, and listed in Table 3, together with tin and lead contents, analytical totals, and the estimated dates or periods. As the results are rather few to draw general conclusions about the relationship between artefact type and composition, the analyses of the later artefacts are included in the survey, and the table has been augmented by the previously published neutron activation analyses of material from Kouklia (Pickles 1988) which had not originally been reviewed in this way. The corresponding catalogue numbers have been prefixed with "NA" to distinguish them from the new material.

The group of bracelets and anklets were from just two sites —the Roman bracelet came from Sheikh Hamad and the other items in the group came from Sa'idiyeh, dated around 1200 B.C. All had reasonably uniform compositions, with 9 to 13 % of tin and 0.5 to 1.5 % lead. Catling (1983) believes that at Lefkandi and Nichoria a high tin content was chosen irrespective of metallurgical

Table 3. Effect of artefact type on composition (Includes some previously published Neutron Activation analyses).

Cat. No.	Period	Artefact Type		Tin (%)	Lead (%)	Total (%)
1	c. 1100 BC	Pendant	Jewellery	0.76	14.16	95.5
2	c. 1100 BC	Fibula	"	2.11	0.27	89.0
7	9th cent. BC	Fibula	"	3.11	0.10	70.7
14	Roman	Fibula	"	2.51	22.9	89.7
5	c. 1100 BC	Pin or nail	"	0	0.06	55.0
6	c. 1100 BC	Pin or nail	"	1.55	0.01	76.0
17	Unknown	Pin or nail	"	3.38	0.17	62.3
26	LC IIIA	Pin or nail	"	0.49	0.001	84.1
11	7th cent. BC	Hook	"	1.10	8.22	95.3
15	Roman	Bracelet	"	9.69	1.10	93.4
30	c. 1200 BC	Anklet	"	9.11	0.83	98.0
31	c. 1200 BC	Anklet	"	12.2	1.11	96.3
32	c. 1200 BC	Anklet	"	10.9	0.92	91.7
33	c. 1200 BC	Bracelet	"	13.3	1.52	95.4
34	c. 1200 BC	Bracelet	"	10.5	0.88	97.2
35	c. 1200 BC	Bracelet	"	10.3	0.56	98.9
36	c. 1200 BC	Bracelet	"	9.62	1.20	98.7
NA 11	LC IIIA	Mirror	"	9.3	-	-
NA 14	LC IIIA	Mirror	"	6.13	-	-
29	497 BC }	Vessel - cauldron	Domestic/ ritual	0.08	0.01	99.3
NA 20	497 BC }	Vessel - cauldron	"	0.07	-	-
37	c. 1200 BC	Vessel - bowl	"	16.3	0.19	80.8
38	c. 1200 BC	Vessel - bowl	"	13.8	0.10	91.9
NA 5	LC IIIA	Vessel - ring based	"	6.1	-	-
NA 6	LC IIIA	Vessel - bowl	"	6.6	-	-
NA 8	LC IIIA	Vessel - bowl	"	11.7	-	-
28	c. 1200 BC }	Tripod	"	1.89	3.77	99.6
NA 18	c. 1200 BC }	Tripod	"	1.82	-	-
16	2 - 3 cent. AD	Pincer	"	0.35	0.17	52.1
24	LC IIIA - IIIA-2	Pincer (tweezers)	"	4.99	0.02	98.0
22	LC IIIA	Chisel	Tools and weapons	7.99	0.03	100.2
23	LC IIIA	Chisel	"	5.27	0.10	98.7
20	c. 1200 BC	Hoe (fragment)	"	8.07	0.02	95.2
21	c. 1200 BC	Adze (fragment)	"	6.88	0.08	76.9
39	c. 1200 BC	Knife or dagger	"	8.13	0.43	86.7
40	c. 1200 BC	Knife or dagger	"	8.68	0.13	96.0
NA 1	LC IIIA	Knife or dagger	"	15.0	-	-
NA 2	LC IIIA	Knife or dagger	"	12.5	-	-
NA 4	LC IIIA	Arrowhead	"	4.6	-	-

NOTE : Neutron activation is incapable of measuring lead, and several other important elements, so no figures are given for "Lead" and "Totals".

considerations to give an alloy with a colour close to that of gold. This concept might have applied throughout the Bronze Ages, because, in the absence of chemical analysis and mechanical testing, colour could be an easy marker for achieving a repeatable alloy composition, as well as being important from decorative considerations (Charles 1980). On this basis, colour might explain the high tin contents of the two mirrors from Kouklia, as a lighter, yellowish colour would be preferable to the dark red-brown of unalloyed copper.

The other items of jewellery, which were rather low in tin with big fluctuations in the concentrations of other elements, all came from the Syrian sites except for a possible pin (**Cat. 27**) from Hala Sultan Tekke, and spanned most of the first millennium B.C. Special mention should be made of the presence of 14.5% zinc in the pendant from Tell Bderi, dated *c.*1100 B.C. Though occasional high zinc contents have been reported from early times, the presence also of a significant amount of tin suggests that the zinc may have been an unknown impurity in the copper as the regular production of brass did not begin till the last century B.C. (Craddock 1980). The Tell Bderi pendant however had less than 1% of tin, but over 14% of lead, and it is difficult to explain this composition except in terms of accidental contamination of the lead by zinc from the smelter flue, or of random experimentation. If the zinc addition were deliberate the motive might have been the achievement of a gold-like colour, as in the alloy "pinchbeck" which contains 7-11 % zinc (Partington 1946). The pendant (**Cat. 1**) had also a metallurgically undesirably high concentration of lead, as did the Roman fibula (**Cat. 14**). These very high lead contents were noted in objects where mechanical strength would not be of paramount importance.

The general category "Domestic/ ritual" includes analyses by both AA and NAA of a fragment of the cauldron from the Persian siege site at Kouklia (497 B.C.) which proved to be almost pure copper. Though Craddock and Gumlia-Mair (1988) have observed that hammered bronzes such as vessels and armour were usually made with 10% tin, pure copper is more malleable than bronze and would be easier to shape into a cauldron if the greater stiffness and the gold colour of bronze were not required. This item does not therefore testify to a tin shortage.

In the case of the tripod, stylistically dated to around 1200 B.C., and analysed by both methods, the 3.8% of lead was almost certainly added to facilitate casting, and is of much greater significance than the small amount of tin. Pincers would be expected to require some hardness to impart springiness; and 5% tin in the 12th century might be reasonable, but almost pure copper in the 2nd-3rd centuries B.C. hints at tin shortage.

In the category "tools and weapons" three sites were represented, all of similar date (*c.* 1200 B.C.). The tin contents were in the range *c.*5% (the arrowhead and one chisel) to as much as 15% for one of the Kouklia knives. Other alloying elements were insignificant. There is every indication that the bronzesmiths understood the merit of avoiding brittleness in impact tools-chisel, hoes and adzes, and striving for maximum hardness in cutting tools. On the other hand, tin contents in the range 6 to 8% are the maximum that can be achieved by adding cassiterite direct to molten copper. Higher tin contents require the addition of pre-smelted metallic tin (Northover 1988).

Arsenic and antimony were detected in some of the unidentified Sheikh Hamad items, but this probably indicates tin substitution rather than preferred use of these alloying metals for a particular type of object.

THE GENESIS OF THE IRON AGE

The first archaeological iron

Iron objects have been reported from contexts dated as early as the fifth millennium B.C., and inventories of pre-Iron Age finds have been published by Wainwright (1936), Coghlan (1956), Forbes (1971, 1972), Wertime (1973), Waldbaum (1978, 1980), McNutt (1990) and Sherratt (1994). For most of the fifth, fourth and third millennia iron appears to have been a very rare and precious metal. It is highly probable that all the earliest iron was meteoritic in origin (Zimmer 1916), and the claim that the "chisel" from Grave A at Samara (dated to *c.* 5000 B.C.) (Herzfeld 1980) was smelted must be viewed with some scepticism since smelting, even of copper, may have been unknown at that date.

Meteoritic iron

Although 93% of all meteorites arriving on the earth's surface are iron-free, 1.5% are of the type known as "stony irons" and 5.7% are classed as "irons" and consist almost entirely of iron-nickel alloys. Of these the type known as "hexahedrites" contain less than 6% nickel as the bimetallic phase *kamacite* whilst "octahedrites" with more than 6% nickel contain both *kamacite* and *taenite* . Both types exhibit a characteristic "Widmanstätten" microcrystalline structure making identification easy, but a third sub-group of "iron" meteorites ("aloxites"), though containing nickel, does not have this distinctive micro-structure. The "stony iron" meteorites have mineral and metallic layers, the latter consisting of kamacite only or kamacite plus taenite, depending on the nickel content. Part of a stony iron meteorite was found on Crete in a Late Bronze Age archaeological context (Waldbaum 1978).

The presence of a Widmanstätten microstructure is a strong pointer to a meteoritic origin but it is not unambiguous and signs of such a structure can be found in smelted iron made from some high-nickel ores. It is also possible that some very early finds reported as smelted might have been made from a meteorite of the aloxite group. The most certain evidence of a smelted origin, revealed by scanning electron microscopy, is the presence of fine bands of silicate slag (Piaskowski 1982), but this examination involves destructive sampling and is rarely carried out.

In Middle and Late Bronze Age contexts occurrences of iron become increasingly frequent, and though claims of a smelted origin have been made for a few of the small number of iron artefacts which have received metallographic examination, the iron objects in the tomb of Tutankhamun (Carter 1972) around 1350 B.C. have been identified as of meteoritic origin.

Textual evidence

References to "iron" in early Near Eastern texts are fairly numerous, though paradoxically, corresponding artefactual evidence for the metal is quite rare. Unfortunately, uncertainties in translation, particularly with respect to place names and to the words for individual metals demand caution in the use of such texts. While McKerrell (1977) proposed that the Akkadian word *annaku* refers to a highly arsenic-enriched alloy, Muhly (1980) insists that the word means tin. Hallo and Simpson (1971) proposed that the word *an-na,* usually accepted as tin, might sometimes refer to arsenic, lead or iron, and Muhly (1973) suggested that the word usually translated as copper or bronze might actually be the word for ice. References to iron in Hittite and Egyptian texts as "Black iron of heaven" and "iron of heaven" tend to endorse the belief in a meteoritic origin for iron up to the mid-second millennium. Two Akkadian words associated with iron –*amûtu* and *asi'u* were translated by Bjorkmann (1973) as meteoritic iron, but Rachel Maxwell-Hyslop (1972) equated the latter with iron ore and *amûtu* with bloomery iron —the primary smelting product— on the basis of Kültepe text CCT4,4a, which refers to a weight loss of 4 shekels in forging a bar with a finished weight of only $^2/_3$ shekels. If however the *amûtu* were meteoritic iron, but of the stony iron type, a 14% yield would be quite reasonable.The material thus produced was stated to have 8 times the value of gold, a price which would be more understandable for a high-nickel, silvery, corrosion-resistant meteoritic steel than for a smelted soft iron.

Some thousand years later, classical writers (e.g. Pliny, *Historia naturalis*) believed that the earliest iron in the world came from the area around the western shores of the Black Sea, with its magnetite-rich self-fluxing black sands. This Chalybean steel contained from 5 to 20 % of nickel, was austenitic, with a silvery colour and good corrosion resistance, and had a hardness of 250-350 HV (Piaskowski 1991). This material would not have been easy to smelt or forge, as evidenced by the slags and residues at the Hellenistic settlement of Petres (NW Greece) where the lateritic iron ores had yielded slags containing prills of nickel/iron alloys and a shapeless lump of iron with 2.25% nickel which Photos, Tylecote and Adam-Veleni (1988) were unable to forge. Nevertheless, since the ancient metallurgists had taken the trouble to bring the lateritic ore from some distance they were presumably able to forge such alloys, as were the early users of meteoritic steel.

Perhaps the most famous ancient text relating to iron is one from the Hittite King Hattusilis III, probably to Shalamaneser I of Assyria at about 1250 B.C. In view of the importance that has been given to this text it seems worthwhile to quote the full translation of the relevant passage:

KBo I 14

20 *As for the good iron*[1] *which you wrote me about, good iron in Kizzuwatna*

21 *in my seal-house is not available. That it is a bad time for producing iron*

22 *I have written. (But) they will produce good iron; so far they will not have finished*

23 *When they have finished, I shall send (it) to you. Today now*

24 *I have an iron dagger blade brought on its way to you*

(Goetze 1940).

On the sole basis of this document has been built the legend that the Hittites were the first to smelt iron and had a monopoly of iron production till the fall of their empire around 1200 B.C. (Zaccagnini 1970).

An alternative interpretation of the Hattusilis letter might be that the Hittites were producing nickel steel, either from meteorites or from the ores of Chalybea, but with difficulty, and in such small quantities that a dagger blade could be a worthy gift for a king. If the document refers to the smelting of ordinary soft iron it implies that the process was not fully understood, and was still a hit and miss affair.

Until fairly recently there was little hard evidence for the presence of iron in Hittite territory, but new work is modifying this picture (Bilgi 1991; Kosak 1986; Muhly *et al.* 1985), though textual evidence remains more abundant than physical evidence.

Smelting and the establishment of iron-working

The ores from which the common metals are produced are mostly oxides, carbonates, or sulphides, associated with varying quantities of minerals such as silica, alumina or limestone. To separate the mineral impurities as low melting point slags, appropriate fluxes are added. When smelting the oxide and carbonate ores of copper the fluxes may be manganese or iron oxides in the form of umbers, ochres or haematite. These combine with silicaceous impurities to form low melting point slags of manganese or iron silicates (fayalite). Mechanical enrichment (Merkel 1985) aided by the bright colours of azurite and malachite, and possibly hydro-refining (Koucky and Steinberg 1982b) meant that high grade ore could be presented to the smelter, and relatively small amounts of slags were produced.

Copper sulphide ores are often associated with an equal amount of iron sulphide, as chalcopyrite. In the matte process a preliminary oxidising roast removes much of the sulphur leaving the copper as cuprous sulphide and the iron as ferrous oxide, which forms a slag of fayalite (ferrous silicate) in the subsequent smelting with silica flux. If the oxidising were over-done the iron could be converted to ferric oxide, which does not form fayalite. (This result is essential in the smelting of iron sulphide ores, to prevent loss of iron to the slag. Here, silica is not helpful, and limestone is usually added to fix the silica as calcium silicate).

The potential exists in both copper and lead[2] smelting for the accidental production of iron if any of the factors silica/ore ratio, matte control, furnace temperatures and reducing atmospheres are incorrect. The eventual discovery of iron smelting by this route was therefore inevitable (Coghlan 1956; Wertime 1980). However, the iron would be intimately associated with gangue as a spongy mass (Clough 1987), from which it would have to be separated by repeatedly heating and hammering to extrude the gangue and produce what is known as bloomery iron.

Considerable time may have elapsed before it was recognised that the soft, dark, corrodable

1. The exact meaning of *parzillu damqu* is unknown; it is not impossible that it denotes some definite quality of the metal.
2. Wertime (1980) records finding a 3000 lb ball of iron near a traditional lead smelter in Iran. The smelter had later been converted into a meachanised blast furnace. He regarded this find as evidence for the discovery of iron as an acidental by-product of lead smelting, but Dennis (1965), describing modern lead smelting practice, notes that scrap iron is added as a flux.

metal thus produced was basically the same material as the silvery, hard, corrosion-resistant nickel iron alloy obtained from meteorites —or possibly by smelting the Black Sea sands with chloanthite (an iron-nickel-cobalt-arsenic sulphide) to give Chalybean steel. Archaeological traces of unambiguously smelted soft iron throughout most of the second millennium B.C. are very sparce and the value of iron indicated by textual evidence was so high that it would seem no serious attempt was made to produce the metal systematically.

MOTIVES FOR THE SWITCH TO IRON TECHNOLOGY

Tin shortage

The source of tin used in the Eastern Mediterranean during the bronze and iron ages has long been shrouded in mystery and much has been written on the subject (e.g. Cleuziou and Berthoud 1982; Crawford 1974; Franklin *et al.* 1977; Mellaart 1976; Muhly 1993; Wheeler *et al.* 1977; Wertime 1977; Yener 1986). Similarly, the trade routes by which the tin reached the bronze producers has exercised many minds (e.g. Muhly 1982). In the troubled times at the end of the Late Bronze Age these might be expected to have a great potential for disruption, and the hypothesis favoured by Snodgrass (1971, 1980, 1981a) that difficulties in the procurement of tin forced the substitution of iron for bronze and initiated the start of the Iron Age would seem to have much to commend it.

Earlier work carried out to test the tin shortage hypothesis (Pickles 1988) analysed material from one just area of Western Cyprus and also considered the few relevant published analyses, but the results indicated that there was no evidence for a tin shortage. The work reported here extends the area of study to bronzes from South-eastern Cyprus, Syria, Jordan and the Cape Gelidonya shipwreck, but the survey has been very far from exhaustive, notable deficiencies being the lack of new material from Greece, the Aegean and Late and post— Hittite Anatolia. Nevertheless, the new results endorse the pattern of previously published analyses of comparable material (Balthazar 1986; Buchholz 1967, 1977, 1982, Junghans *et al.* 1968; Peltenburg 1981; Rapp 1981; Stoss-Gale *et al.*1986; Swiny 1986; and Weinstein 1980). and support the belief that around the end of the 13th and the beginning of the 12th centuries B.C. there were adequate supplies of tin in the areas studied with the exception of Northern Mesopotamia (Moorey 1985).

The statistics of archaeological finds of all types of bronzes for this period indicate a continuing and even increasing use of bronze during what is regarded as the Early Iron Age. Thus, the compilations of Waldbaum (1978), Buchholz (1969) and McNutt (1990) show that bronze tools and weapons greatly outnumbered those of iron in all the areas of the Aegean and the Near East during the 12th and 11th centuries B.C., by which time the iron industry could be said to be established though not yet dominant (Snodgrass Stage 2). Even during the 10th century bronze finds outnumbered those of iron in all the areas except Palestine. In Cyprus by the 10th century and perhaps the 9th century in most other areas iron was beginning to overtake bronze for utilitarian objects (Snodgrass Stage 3), but bronze continued to be used in abundance for those purposes for which it was more suitable and there appears to have been an overall increase in the use of metal. Thus, in Luristan, where the first evidence for iron usage was in Iron II (900/800 to *c.*750 B.C.), bronze continued to dominate and was still used for weapons as well as for ornaments in Iron III (8th to 7th centuries B.C.) when iron had become very common (Muscarella 1988a and b; Van den Berghe 1968).[3]

Many of the data come from early excavations which did not have the benefit of modern skills

3. It bears repeating that statistics of archaeological finds must be treated with caution. Excavated sites represent only a tiny and random faction of the occupied regions of the ancient world, and even the most extensive campaigns rarely excavated more than a small part of the whole site. The pattern of excavation is strongly influenced by non-archaeological considerations, for example the need of rescue excavations where major public works are planned, the driving force of Biblical research for work in Palestine and Israel, and the helpful attitude of the Cypriot authorities which has drawn many foreign archaeologists to their island.

and techniques. Iron objects corrode rapidly into swollen masses of rust which may have been ignored in earlier times when the acquisition of exhibits for museums and private collections was the prime objective of archaeologists. This would distort the statistic of iron finds unless only the more recent and reliable excavations are considered. On the other hand, bronze, unlike iron, is easily recycled, so the number of objects deposited (either deliberately or accidentally) was probably only a small proportion of the total in circulation over a long period. A further complication is the preponderance of finds from burial rather than occupation sites, which distorts the statistics of artefact types, diminishing the percentage of tools and utilitarian objects (which in any case would have a high chance of being recycled) in favour of jewellery and prestige items.

Dating[4] also presents difficulties. There is no known scientific method comparable to radiocarbon or thermoluminescence dating which can be applied to metals, and even if the context of discovery can be dated unambiguously, the item may have been of some antiquity at the time of deposition (There is however a possibility of dating metallurgical slags by thermoluminescence methods, or by carbon dating of charcoal inclusions.)

Far more significant than the random statistic of numbers of finds is the pattern of bronze analyses at this time, which shows no reduction in the use of tin. In this work especially, the compositions of the Sa'idiyeh material, where bronze and iron shared the same chronological context, indicate ready availability of tin for new bronze manufacture (e.g. not incorporating scrap bronze). It is disappointing that no bronzes in category C (where bronze objects and iron weapons were found in the same grave) were suitable for analysis, but it is hoped that XRF analyses at present in progress at the British Museum laboratories (Cowell pers comm) on some of the corroded artefacts from Sa'idiyeh will provide qualitative information about their compositions.

It must therefore be concluded that an interruption in tin supplies was not a prime cause of the escalation of iron production at the end of the Late Bronze Age. It is interesting to note that the analyses of bronzes in this work from the later Syrian sites, when the Iron Age was well established, show no signs that tin was more freely available in that region than throughout most of the second millennium B.C.

Fuel shortage as the trigger for the Iron Age

The theme of deforestation, in which fuel-hungry metallurgical processes competed with injudicious over-grazing was long a concern of Wertime[5] (1983) and he believed that a major reason for the transition from Bronze to Iron Age was because iron smelting consumed much less fuel than bronze smelting (e.g. Horne 1982) and that denudation of the forests forced a shift to a less fuel-hungry process (Constantinou 1982; Wertime 1980, 1982). The evidence for this was examined by Waldbaum (1989), who was not fully convinced. Though there is evidence of deforestation resulting from intensive metallurgical production, as at Pithikousai in the 9th to the 8th centuries B.C., the culprit was iron, and the period after the Bronze/Iron Age transition. The Etruscan iron industry was moved by the Romans to the mainland because of deforestation, whilst in Siegerland (Germany) iron smelting was restricted to three months of the year (to take advantage of coppicing) because of earlier deforestation (Weisgerber 1981). In Cyprus on the other hand, the problem appears to have been one of excessive afforestation, if the writings of Strabo quoting Eratosthenes are correctly dated to the 8th to 7th centuries B.C.

The claim that iron smelting was more energy-efficient than copper smelting seems to fly in the face of thermodynamic data if one considers the heats of reaction for the main chemical processes in copper and iron smelting. Considering first the reduction of the oxide ores the following, equations apply:-

4. In this work, radiocarbon dated adjusted according to the MASCA calibration have been used where possible.
5. Despite this conviction, Wertime himself (1982) stated that copper and bronze production peaked in Western Asia (including Cyprus and Egypt), and also in China and in Europe, *after* the iron age was well under way.

OXIDE ORES

COPPER

(1)	$Cu_2O + CO$	=	$2Cu + CO_2$	(Heat emitted = 13.9 K.cal./gram atom copper)
(2)	$CuO + CO$	=	$Cu + CO_2$	(Heat emitted = 30.7 K.cal./gram atom copper)

IRON (Reactions typical of a modern blast furnace)

(3)	$3Fe_2O_3 + CO$	=	$2Fe_3O_4 + CO_2$	(Heat emitted = 2.3 K.cal./gram atom iron)
(4)	$Fe_3O_4 + CO$	=	$3FeO + CO_2$	(Heat absorbed = 2.8 K.cal./gram atom iron)
(5a)	$FeO + CO$	=	$Fe + CO_2$	(Heat emitted = 3 K.cal./gram atom iron)
(5b)	$FeO + C$	=	$Fe + CO$	(Heat absorbed = 40.0 K.cal./gram atom iron)

The reduction of ferrous oxide by carbon (5b) accounts for only about 10% of the total, but it brings the net heat balance for reactions (5a & b) to a small absorption of less than 1 K.cal./g.atom of iron. Since the normal starting material is haematite (Fe_2O_3), there is a total heat absorption of about 0.35 K.cal./gram atom of iron. The addition of limestone as a flux also adds to the fuel consumption by the reaction:-

(6)	$CaCO_3$	=	$CaO + CO_2$	(Heat absorbed = 42 K.cal./mole).

SULPHIDE ORES

COPPER

(Iron sulphide is rarely used for dedicated iron smelting, but the chemistry follows the equations 7b and c and 8 b below. Instead of reaction 8a, silica is removed by combination with calcium oxide.)

Chalcopyrite with silica flux. Note; Chalcopyrite is effectively iron pyrite (FeS_2) in which some of the iron atoms have been replaced by copper. For clarity, the ore has been represented as an equimolecular mixture of the two sulphides.

(7a)	$^1/_2CuS_2 + 1^1/_2 O_2$	=	$^1/_2Cu_2S + 1^1/_2 SO_2$	(Heat emitted = 73.5 K.cal./ g.atom Cu)
(7b)	$FeS_2 + O_2$	=	$Fe S + SO_2$	(Heat emitted = 51.2 K.cal./g.atom Fe)
(7c)	$FeS + 1^1/_2O_2$	=	$FeO + SO_2$	(Heat emited = 113.6 K.cal./g/atom Fe)
(8a)	$2FeO + SiO_2$	=	Fe_2SiO_4 (fayalite)	(Thermodynamic data unavailable)
(8b)	$FeO + CO$	=	$Fe + CO_2$	(Heat emitted = 3 K.cal./g.atom Fe)
(9a)	$2Cu_2S + 3O_2$	=	$2Cu_2O + 2SO_2$	(Heat emitted = 46 K.cal./atom Cu)
(9b)	$2Cu_2O + Cu_2S$	=	$3Cu + SO_2$	(Heat absorbed = 5 K.cal./g.atom Cu)
(9c)	$Cu_2O + CO$	=	$Cu + CO_2$	(Heat emitted = 14 K.cal/g.atom Cu).

Reactions 7a, b and c and 8a take place in the *matte* stage of copper smelting, although in ancient times the initial roasting may not have proceeded beyond 7a and b. Reactions 9a, b and c take place in the main smelting stage, where the net heat emission is between 41 and 60 K.cal./ gram atom of copper, depending on the division of the reduction between reactions 9b and c. The total heat balance of the various reactions including the matte stage indicates a net heat evolution of 140 to 149 K.cal. per mole of $(Cu,Fe)S_2$, or 279 to 298 K.cal./g.atom copper. (The oxidation of the iron in the chalcopyrites makes a large contribution to the total heat evolution, but most of this is lost in the *matte* stage.)

Thus, considering only the chemistry of the processes, the smelting of copper sulphide ores is accompanied by the emission of heat, whilst that of iron oxide ores requires a net input of heat.

The main arguments for the high fuel requirements of copper smelting appear to be based on the assumption that slag viscocities would be very high, and that the furnace temperature would have to be maintained at maximum for a long time to allow the copper to percolate through. However, if the composition of the charge was correct for converting all the iron and silica to fayalite the slag produced would be very fluid, and the difficulty would not arise (Bamberger 1985). An imbalance would give inhomogeneous and more viscous, mushy slag which could retain metallic copper as prills. With a deficiency of silica, iron could be reduced to metal in the zone below 700 °C.

Though such slags (usually not tapped, and of Koucky's "C" type: Koucky and Steinberg 1982a) are well known we do not believe that the main production of copper depended on its mechanical recovery from these residues.

Some investigations have been made of the relics of early smelting furnaces. For example Rothenberg, Tylecote and Boydell (1978) at the early mining site of Timna found evidence of copper-working, both mining and smelting, in the Chalcolithic[6] and Early Bronze Ages. The slags associated with a simple bowl furnace of the 4th millennium B.C. indicated the use of carbonate/oxide ores (malachite and paracatamite), requiring iron or manganese oxide flux. The slags had high viscosities and high melting points of 1180-1350°C and contained entrapped copper prills, as described above. However, a primitive stone-built furnace of EBA II date showed certain evidence for the tapping of the slag, indicative of higher operating temperatures, a more fluid slag and better segregation of the copper.

Smelting experiments were carried out in reconstructions of the furnaces (Rothenberg 1978, 1985, 1990), the most successful being the interpretation of the chalcolithic furnace, using Timna ore. A 95% yield of copper was obtained by adding the ore mixed with half its weight of charcoal to a charcoal-filled furnace pre-heated for 21 hours. The overall charcoal consumption (including pre —and post— heating) was 6 .64: 1, a far cry from the 300: 1 quoted by Constantinou and the 620: 1 attributed by Wertime (1982) to Tylecote (no ref. given). (N.B. Tylecote (1980) specifies a 40: 1 fuel: ore ratio for copper sulphide smelting.)

The Timna experiments used a single, inclined tuyere, but much better results were obtained by Bamberger (1985) using up to six tuyeres, to give better air distribution and enlarge the reaction zone. With $1^1/_2$ hours pre-heating and the same (2:1) ore to charcoal mixture the copper melted through a very liquid fayalite slag and formed a 1.5cm. thick plate beneath a 4cm. thick slag cake.

Unfortunately, most of the published scientific examinations of slags do not record the melting points, which could give valuable information about furnace conditions. The chemical compositions show a preponderance fayalite, with occasional wüstite (FeO), and numerous impurities including calcium, aluminium, iron and copper. For tapped slags the furnace temperatures must have been significantly higher than the slag melting points, which for fayalite is 1205°C (Bowen and Shairer 1932). There is a range of compositions for the iron-silicon-oxygen system melting below 1300°C, including two eutectics melting at 1177 and 1178 respectively. It seems likely that maximum furnace temperatures reached at least 1300°C. These temperatures were probably reached even in the earlier crucible furnaces if we are to reconcile the "C" classification of Late Bronze Age slags (Koucky and Steinberg 1982a) with the high quality artefacts associated with the period. It should be noted that once iron is converted to silicate it cannot be smelted to metal without roasting with lime (calcium oxide or carbonate), though a very small amount of iron can be recovered by re-melting fayalite as a result of partial disproportionation.

These observations show that it would have been possible for equally good results to have been obtained in the past, using equipment consistent with the surviving evidence. Perhaps insufficient allowance is made for the intelligence and experience of the Bronze Age metallurgists, built up over thousands of years. It is difficult to believe that the scale and quality of bronze production indicated by archaeological discoveries could have been achieved by such primitive technology as is often assumed. Evidence that, by the Late Bronze Age at least, copper smelting had reached a much more advanced stage than is sometimes assumed from the sparse archaeological remains of smelting furnaces is provided by the copper oxhide ingots (e.g. Gale and Stoss-Gale 1985; Muhly, Maddin and Stech 1988; Muhly, Maddin and Wheeler 1980) which were widely traded at that period. Tylecote (1980) considers that these

6. Muhly (1984) disputes the evidence for mining or smelting at Timna before the 13th century B.C.

massive items, weighing 30 to 40 kilograms, could only have been made by tapping the whole charge of a smelting furnace direct into the mould. The sophisticated nature of LBA Cypriot bronze industry is demonstrated by the many technologically advanced cauldrons, tripods, and especially by the famous 'Horned God of Enkomi' (Dikaios 1962).

It is difficult to find authoritative figures for the consumption of charcoal in early iron smelting. In contrast to the views of Wertime (1983), Horne (1982) and Constantinou (1982), Dever (1992) specifies a fuel consumption for iron production five times greater than that for bronze (no authority quoted). Nielson (1987), based on West African field work and on a study of Etruscan and Roman slags, deduced that six tons of charcoal would be needed to produce 200 kilograms of iron – very much worse than for copper. A modern blast furnace, with thermal recycling and the advantages of size and insulation and continuous operation consumes about 0.85 tons of coke per ton of iron (Dennis 1965), but a Polish reconstruction based on archaeological evidence of a Roman iron smelter, required a charge with a charcoal to ore ratio of 2:1 to be added to a furnace which had been pre-heated for 18 hours (Nosek 1985). After smelting, the iron bloom had to be consolidated and shaped by repeated heating and hammering in a thermally inefficient forge, a process which must have consumed a considerable amount of fuel.

The need for pre-heating is common to both copper and iron smelting, though in modern continuous smelters this fuel overhead is spread normally over the operating life of the smelter. In ancient times pre-heating probably accounted for the largest part of the fuel consumption, so it is instructive to compare the 1½ hours required in Bamberger's experiments with the 18 hours of the Polish iron smelting experiments. The actual fuel consumption in the ancient metallurgical processes must have been determined predominantly by the design and scale of the furnaces and the efficiencies of the technologies.

The evidence clearly does not justify claims that the smelting of copper was greatly less fuel-efficient than that of iron, and that the change to iron as the main material for tools and weapons was dictated by the need to conserve fuel. There is in fact no evidence for severe timber shortage in the 13th-12th centuries B.C. in the metal-producing regions of the Near East. On the other hand, the vast increase in metal production once the Iron Age was well under way, made possible by the abundance and widespread distribution of iron ores, was perhaps the greatest single cause of deforestation since the neolithic revolution.

Carburisation as the trigger for the full Iron Age

Forged bloomery iron is a soft metal, greatly inferior to a worked bronze for cutting tools, and would be acceptable only if bronze were unavailable or very much more costly. Conversion of the wrought iron to steel by the process of carburisation and quenching increases the hardness dramatically, and produces a material of superior cutting properties and great desirability (Maddin 1981). Thus, it is possible that the first use of iron for specifically domestic purposes may have been the production of one-sided knives in Cyprus, from the end of the Late Bronze Age. These appear to have been carburised and probably quenched, achieving a hardness greater than that of bronze (Tholander 1971). Though Constantinou (1982) is dismissive of the Cypriot iron resources because of the lack of haematite deposits, the use of chalcopyrite in copper smelting throughout the Late Bronze Age in Cyprus gave great potential for the accidental or deliberate production of iron, as has been pointed out earlier. Iron ore (probably iron oxide gossan) was apparently mined in at least two places in Cyprus, according to Cobham (1908) quoted by Kassianidou (1994). She shows that the much-quoted statement by Steinberg and Koucky (1974) that Cypro-Phoenecian slags from which iron could be isolated by hammering were sold by the Ottomans for re-smelting was a misinterpretation of the comment by Richard Pococke, 1738.

The Cypriot knives were a great success and appear to have been exported to or copied in Palestine and the Levant (Waldbaum 1980). The secret of this success was surely the hardness achieved by carburisation and quenching. As this process requires prolonged contact with carbon at high temperatures it is difficult to achieve in a conventional open forge, but immersion of these small objects in the upper hot zone of a deep furnace where the temperature had fallen to below *c.*1000° K could speed up the carburisation process as a result of the deposition of atomic carbon on the metal by the reaction:-

(10) $2CO = C+CO_2$ (Heat emitted = 18.7 K.cal./mole CO)

This reaction is possible in the absence of oxygen because carbon dioxide is more stable than carbon monoxide at lower temperatures. Above about 1000° K carbon monoxide is more stable and the reaction proceeds in the reverse direction. In an actual furnace, the main reaction in the lower zone is the combustion of carbon to its oxides, with the production of heat. In this zone the temperature may exceed 1650° K and any oxygen and carbon dioxide in the blast are rapidly converted to carbon monoxide. Above this zone when all the oxygen has been used up the production of heat ceases and the temperature gradually falls. As a result there is a region where Boudouard's reaction (Equation 10) is possible, and this has been shown to take place in the upper/middle region of a blast furnace (Partington 1946). The reaction is also known to cause degradation of catalysts in the exhausts of internal combustion engines (Tavares *et al.* 1944). A note on the thermodynamics of the reactions is given in the Appendix.

In a typical blacksmith's forge the metal is placed on the surface of the hot bed of fuel where there is sufficient access of air to prevent the deposition of atomic carbon (Piaskowski 1991). This would also apply at the top of a deep smelting furnace, but if in the forging of their knife blades the Cypriot smiths chose rather to immerse them deep in the hot fuel bed, deposition of atomic carbon by Boudouard's reaction could have led fortuitously to the production of carburised steel. Subsequent quench-hardening could have been the chance result of a wish to cool the knives rapidly for handling purpose. Their small size would make it easy to drop them in a bowl of water. Unfortunately, detection of tempering, which would provide strong evidence that the process of quench hardening was deliberate and understood, relies on the observation of remnant pseudo-martensite structures which are difficult to detect, being often largely destroyed by corrosion (Knox 1963; Smith 1967). Carburisation was detected in the bangles and items of jewellery from the Timna excavations (McGovern 1986) but the steel formation may have been accidental (Clough 1987), again perhaps the result of deep immersion in the forge.

Many of the earlier knives incorporated bronze rivets to secure the handles (Waldbaum 1982); out of 43 listed by Sherratt (1994) from twelfth century contexts, 20 had bronze rivets, compared with 19 out of 81 for the eleventh century. Sherratt has interpreted this trend as reflecting the declining prestige of the knives, but an alternative explanation could lie in the method of making the rivets. If originally the same process was used as for the blades, iron rivets would have been too brittle and the ductility of copper or bronze would be sought. Later, when the carburisation/quench hardening and tempering process was more fully understood it would be possible to produce suitably ductile iron rivets.

It is not really possible to say when and where the first iron smelting took place —the Kerman range has been suggested (Caldwell and Shahmirgadi 1966), while Tubb (1988b) quotes Gordon (unpublished) for evidence of very early iron smelting at Yelul edh-Dhabab on Wadi Zerqa, Jordan. The origins of the process of converting soft bloomery or wrought iron into steel by adjustment of the carbon content are equally obscure, and though several isolated occurrences of carburised steel are known (e.g. the five anklets from the Baq'ah Valley in Transjordan (Piggot *et al.* 1982) which appear to have been produced accidentally, and unmodified by subsequent heat treatment), there is a good case for postulating that carburisation was first systematically applied in Cyprus, and that the Cypriot iron knife industry from about 1200 B.C. marked the start of Stage 2 of the Iron Age in the Eastern Mediterranean according to Snodgrass' classification.

Although carburisation would not be so easy to achieve with large objects, the wide distribution of the steeled knives would spread knowledge of iron as a cheap and readily available metal for many uses, not just as a substitute for bronze. It is perhaps significant that though Cyprus may have the first centre for the manufacture of every-day iron objects, the production of bronze —which came from the same ore— continued to dominate the local metal industry, whilst on the Asian mainland over the next three centuries a large and thriving iron industry developed, based on the abundant deposits of haematite ore which Cyprus lacked.

The important part that Cyprus appears to have played in changing the status of iron from that of a rare and prestigious material to that of a cheap and plentiful metal for numerous utilitarian purposes may be a consequence of the nature of Cypriot copper ores, coupled with the chance discovery of carburisation and quenching. By contrast, Timna, which had a long history of copper metallurgy from Chalcolithic to Late Bronze Age based on oxide ores, and was an important iron-working centre later in the Iron Age, appears to have lagged behind Cyprus in the introduction of iron technology.

If carburisation and quenching marked Stage 2 of the Iron Age and created a wide knowledge of and demand for iron (Stage 3), it would seem that knowledge and understanding of the process was not disseminated as widely and successfully as that of the basic production technique for iron, since the proportion of carburised objects declines in subsequent centuries. For instance, at Kinneret (Muhly *et al.* 1990) most of the iron objects from the 11th and 10th centuries had evidence of carburisation while most of those from the 8th century did not. This may be because of the difficulty of carburising large items, and a failure to realise that total immersion in a deep charcoal bed for a prolonged period was needed.

THE BRONZE/IRON TRANSITION IN CONTEXT

This study provides additional evidence against tin shortage or fuel depletion as adequate reasons, either jointly or singly, for the development of iron. It also reaffirms the pre-eminence of Cyprus in the inauguration of Stage 2 of iron-working. Although unproven, it seems likely that this Cypriot innovation was adopted by regions within the Cypriot interaction sphere. While its pivotal role in copper production and distribution in the Mediterranean prior to *c.* 1200 B.C. is well known, the post-Sea Peoples phase is often regarded as one of relative isolation and decline. Recent studies have shown that this view is no longer tenable and that vibrant trade ensued in the Early Iron Age (Muhly 1992; Karageorghis 1994). Thus, Cyprus possessed the means, motive and opportunity for the dissemination of novel metal products within the altered social and economic conditions of the 12-11th centuries B.C. But that does not provide satisfactory insights into reasons for the innovation. Susan Sherratt attempts to do this in a general manner by re-thinking the socio-economic circumstances surrounding the emergence of iron. She emphasises the absence of a centralised political system in Cyprus, the existence of free-lance seaborne trade alongside regulated exchange, urban control of production and product replacements in the 13th century B.C., intensifying diversification of metalwork in the 13th-12th centuries leading to subversion of the established order and the existence in Cyprus of value-added products and commercial trade or marketing strategies aimed largely at sub-elite markets (Sherratt 1994, 1998; see also Muhly 1992, 19).

Table 4. Reported occurrences of copperworking evidence at Enkomi. Qartier 1W: shaded bar represents varied installations, debris and other copperworking evidence; open bar, little evidence.

LC Period	*Town*						*Qartier 1W*
	Smithy	Tuyere	Crucible	Mould	Scrap	Slag	
IIIB	•••	•	•	•	•••		
IIIA	••••••	•	•	••	•••	•	*Houses*
IIC	••			••			
IIB							
IIA							*Fortress*
IB							
IA			•			•	

We seek to approach this breakthrough from a related, but slightly more focused perspective, namely the contextual analysis of evolving practices that occurred in a sequence of copper and bronze-working locales. We do this on the assumption that iron was regularly encountered in the course of smelting and refining the copper (Charles 1980; Wertime 1980; Gale *et al.* 1990), where iron was initially present in the ore as chalcopyrite or added as a flux for copper oxide ores, typically ochres or gossans, as argued above and by others (recently: Sherratt 1994, 66; Snodgrass 1994, 168). The formation of metallic iron in such circumstances would be commonplace, as demonstrated by the presence of up to 10% of iron in Cape Gelidonya copper ingots (Maddin and Muhly 1974). Muhly opined that copper slag may have been the source of iron, but Maddin remained equivocal (1982, 311). We doubt that old slags were re-smelted to supply iron (the chemistry is against it), but we believe that the metal may have been produced as a by-product of copper scavenging in the course of current smelting operations. In a more recent discussion, Kassianidou effectively dismissed the slag hypothesis but allows that iron compounds added as fluxes for working copper may have been reduced accidentally to metallic iron (Kassianidou 1994; see above for the situation at Hala Sultan Tekke). In the absence of evidence to the contrary, we can only state that copper smelting seems to us to remain a plausible source for early iron.

According to Sherratt (1994, 86-7) two precocious centres for early ironworking may have been Palaepaphos and Enkomi. Of these, only Enkomi has the chronological depth and spatial breadth required by this approach.

Thanks to Dikaios' brilliant work in Enkomi Qartier 1W, the area of the Fortress (also known as the industrial complex), we have a controlled stratigraphic sequence of copperworking that spans most of the Late Bronze and Early Iron Age (Dikaios 1969, 1971). During the LC I-IIB periods it seems that most of the copper was processed in the Fortress smithies (see Table 4). This coincides with the time when the building physically dominated the town, suggesting the existence of an overarching authority, probably resident in its upper stories. In order to assure the smooth flow of copper from the mines, it controlled a three-tier settlement/mining hierarchy in a territory that stretched to the Troodos copper sources (Peltenburg 1996, 30-35) and it probably attracted an ever-increasing population from the surrounding country (Keswani 1996) by virtue of its gateway status. Although not the only exporter of copper, the wealth of exotics in its tombs, the sustained evidence for large-scale copper production and the overwhelming eastern Cypriot character of pottery exports to sites like Tell el-Dab'a point to its pre-eminence in Cyprus at this time. Knapp argues for many more copper-producing centres (Knapp 1986, 73-4) and Keswani for more widespread occurrence of valuables in tombs (Keswani 1996, 229), but much of the quoted evidence is used retrospectively. It belongs more properly to the 14th-12th centuries B.C. when the situation was different.

Production residues at the Fortress peaked in the 13th century B.C. Dikaios recorded several smithies with varied evidence for metalworking and a large, $80m^2$, slag dump outside its west wall adjacent to a concentration of workshops (Dikaios 1969, 46-66). Although he was at pains to correlate this with the influx of Aegeans (Dikaios 1971, 512), extensive copperworking already occurred in the preceding level, IIA, where he found *in situ* remains of refining in the west wing and courtyard, equipment such as moulds, a possible ingot fragment and slag (Dikaios 1971, 507-8). Thus, in spite of the possible destruction of IIA, there is continuity of copper production here into the 13th century B.C.

The continuous Levels IIA-B metalworking trajectory masks significant organisational changes that took place within the Fortress. These changes, moreover, are part of wider transformations that accompany the start of LC IIC Cyprus (cf. Knapp 1996, 68). They are evident in other parts of Enkomi, in regional settlement patterns and in the influx of exotica.

At the outset of Level IIB, the rooms of the Fortress were re-arranged to constitute independent units or households (Fig. 2). For some three centuries prior to this time, it was a unified entity framed by thick walls (Level 1A), with interconnecting communications throughout (Level IB) and, latterly, redesigned so it had a row of rooms fronting a 27 m. long courtyard (Level IIA). This coherence was abandoned in the 13th century when the whole area was redesigned to form independent but contiguous structures. In the last phase of IIB, the row of the now independent houses backed on to a newly installed fortification wall. Dikaios seemed aware of this fragmentation when he identified distinct sectors, but then went on to characterise them as "forming part of a continuous whole" (Dikaios 1971, 510). His appreciation was largely based on the argument that a Mycenaean industrialist installed himself here, with megara in the central sector, ladies residences in the east and copper workshops in the west (Dikaios 1969, 66). In fact, the sectors constitute at least four discrete multi-roomed units constructed around core rooms 87, 27/3c, 1/59 and 47, and the recognition of central megara in this assemblage seems to us to be an over-enthusiastic reading of the evidence. As they do not interconnect, it would be more appropriate to describe the Level IIB 'Fortress' as a group of Enkomi Qartier 1W houses. While it is clear that most evidence for metalworking occurred in the western building focused on court 87, residues and installations were found in the two easterly structures as well, hence most houses were engaged in metalworking. This devolved architecture continued into Level IIIA when there is evidence for more subdivisions and accretions in the west. Although walls are scrappy in IIIB and IIIC, there is no return to the much earlier unity of the Fortress. The Level IIB re-arrangement, therefore, was an enduring and fundamental break-up of a dedicated metalworking centre.

In terms of the organisation of metalworking, we may infer that this architectural disintegration corresponds with the inception of a multiplicity of more or less autonomous family businesses specialising in copper production. It is consistent with a process of industrial devolution seen in the wider reaches of 13th century Enkomi.

In 1981, Courtois attempted to demonstrate the long history of metalworking at Enkomi in order to counter the prevailing argument that it only expanded enormously under the aegis of Aegean colonisers in the 12th century B.C. (Cou tois 1982). Using the French records (many c them previously unpublished) and Dikaios' publ cations, he was able to point to the ubiquity c slags, portable metalworking equipment an foundries in the town. He was, however, ham pered by defective chronological controls, distu bance of earlier remains in a non-tell formatio and relative paucity of Level IA and B exposure It is difficult now to obtain a very clear idea of th extent of excavation in the earliest levels, bu Courtois was assiduous in recording all instance of metalworking installations he could find (if nc necessarily of slag). He mentions several Frenc exposures of LC I and his survey includes 120 m.[2] of Level 1A and IB deposits from Dikaios Area I alone (Dikaios 1969, pls 266-70). Grante we are dealing with a biased record in which ea lier deposits are under-represented, the evidenc is consistent for a remarkable shift in the distribu tion of copperworking residues in LC IIC-IIIA There is a transitional phase in LC IIC when rem nants occur in both zones, that is in Qartier 1V and in other quarters of the city, but by LC III the re-location is virtually complete, with onl one of the rooms (56) in Qartier 1W possessin evidence for working (although moulds occu elsewhere) (Dikaios 1971, 516-7). While othe have noted this spatial dichotomy (Courtois 198 157, 160; Keswani 1996, 226), we wish to empha sise the profound organisational shift that precip itated these altered patterns. Against this, it coul be argued that since central authorities controlle elite craft operations in dispersed locations, th re-location is not to be equated with emancipa tion. Mesopotamian palaces, for example, orga ised crafts beyond the physical confines of th palace (e.g. Dalley 1984, 50, 62-3) and the issu of metals from Mycenaean palaces to rural ate liers, as indicated by the Jn series of Pylos Linea B tablets (Hooker 1982), are two Bronze Ag instances of centralised, long-distance contro But evidence for a Cypriot palace economy at th time is weak. The Enkomi metalworking dispe sion, the emergence of houses where previousl the Fortress stood and the contemporaneous pro liferation of newly established prestigious tow houses suggests that LC IIIA represents the cu

Fig. 2. The devolution of an architectural entity. Disaggregation of the components of the Enkomi Fortress occurs in c (Level IIB). a (Level IB), b (Level IIA), c (Level IIB), d (Level IIIA).

mination of a long term trend towards the dissolution of central control of metals.

The replacement or weakening of a central institution at Enkomi, first clearly discerned in Level IIB (=LC IIC), has resonances in other parts of the island. It is about then that new centres spring up where there had been either few signs of previous settlement (e.g. Kition) or where only low levels of political and economic integration are evident (e.g. Vasilikos Valley) (most recentrly: Manning 1998). The emergence of these competing south coast polities is strongly linked to copper production and export. During the 13th century B.C., for example, there is a surge in imports of foreign luxuries like faience and glass, with a concomitant rise in local enterprises, often imitating the imports (cf. glass pomegranate bottles: Peltenburg 1986). Indeed, it is difficult to ascribe any imports of these materials prior to the 13th century in these *nouveaux riche* regions, and other exotica are scarce before then. Taking imported faience vessels as a guideline, it is clear that the character of imports is significantly different from those at Enkomi (Peltenburg 1974, 138-9). From this we may infer separate long-distance trade relations and hence the emergence of independent south coast polities at least by the 13th century. There are, therefore, several reasons for postulating significant decentralisation in Cypriot sociopolitical and economic structures in the late 14th-early 13th century B.C. To judge from the more outspoken profit-led utterances of the king of Alashia (Liverani 1979, 28-9), its roots lie further back within the organisation of Cypriot society. And with the disappearance of palace dominated centres of international networks like Ugarit, the Early Iron Age presented opportunities for the furtherance of decentralised activities at many levels of society. The Sinaranus of the Levant were at last unfettered and, as described in the later report of the hapless Wen-Amon, sea-going freighters independent of the crown plied their trade to various commercial houses (Simpson 1973, 142-55).

But how did this deregulation foster ironworking? And why, if at Enkomi there were alternating cycles of hierarchical political integration and ascendancy of competing elite groups (Keswani 1996, 226), did it not develop earlier? To address the last question first, we have shown above that the Level IIB shift was so pronounced that it was not just one more episode in power fluctuation, but a definitive rupture. Many other factors, including the violence that attended this restructuring process, need to be taken into account, but we believe it is useful to focus more directly on the situation of the independent metalworking houses of Enkomi in these new 13th-12th centuries B.C. circumstances to evaluate possible incentives for the phenomenon.

During the earlier, large-scale centralised operations at Enkomi, we suspect that primary sources of copper ore and matte were relatively assured and that attached metalworkers here or closer to the mines were pressured to deliver pure copper in bulk without being over-zealous in extracting every ounce of the copper from delivered raw materials. In the more competitive environment of LC IIC-IIIA, downsizing ensured each piece of metal became precious, so there is a concomitant intensification of scrap hoarding (at Enkomi) and transport of scrap (Cape Gelidonya wreck). Production may now have been centred on self-employed artisans and family businesses where maximising the yields of saleable products from the available materials would be more important than minimising labour input. One way to achieve enhanced production levels was to expend more effort on extracting copper from iron rich ores and mattes.

In the upper zone of a smelting furnace, conditions favour the reduction of iron oxide by carbon monoxide at temperatures well below the melting point of copper. Any metallic iron so formed will be associated with the infusible material. It will not combine with silica to form fayalite, which is the main component of tapped slags, but will remain recoverable in furnace residues and non-tapped slags. These residues are likely to contain copper prills which could be recovered by repeated heating and hammering, a procedure which would also separate and consolidate any metallic iron in the form of a 'bloom'. Continuation of the same process would convert the hammered flat bloom into wrought iron, not dissimilar in shape from a knife blade. Knowledge of the procedure for forging and shaping may have pre-existed (meteoric iron had been known for some 2000 years) but in any case it would be self-evident from the process by which the iron bloom had been obtained. The amount of iron recovered from a single smelting batch would be relatively small, perhaps just right for making one of the small knives which were the hallmark of the early Cypriot iron industry. The size of these knives would favour full immersion in the furnace during the forging operation, so yielding the carburisation which has been discussed above as one probable cause of the success of the Cypriot knives. Several aspects of this model have recently received striking confirmation from the discovery of co-smelting of copper and iron at Late Bronze Age Tell Brak.

Shell's (1997) studies of this remarkable material prove conclusively that iron was formed during the smelting of copper matte in the 15th-13th centuries B.C. in northern Mesopotamia. Crude copper ingots cast from smelting furnaces contained dendrites of iron typical of those formed on cooling of solid solutions of iron in copper. The high concentrations of iron indicate that the solid solutions had formed at temperatures of 1400 deg. C or more. In the upper, cooler part of the furnace iron forms as copper-rich bloom, and one specimen of such material was found.

A sample of iron-rich silicate slag from the Mitannian palace (HH548) indicates that the crude copper was purified by melting with silica sand rather than by relying on floatation-melting alone to separate the iron.

Further, a possible arrowhead (HH149) of worked iron from a 13th century Middle Assyrian context shows utilisation of the accidentally produced iron.

Clearly, some Near Eastern metalworkers were experimenting with iron production in the course of copper working during the Late Bronze Age. Although we lack relevant material from the Enkomi smithies to ascertain if copperworkers there were definitely engaged in similar practices, our model has two strengths.

First, by highlighting that ironworking is a socially constructed activity it focuses on a sequence of metalwoking contexts rather than on general historical events (*contra* Mirau 1997). Second, the hypothesis can be tested. Analysis of ores, slags and other waste products from a sequence of relevant contexts, especially accessible material from Enkomi, might confirm the assumptions about the types of ores and fluxes in use. An absence of copper, especially of prills, in slags and furnace residues might be indicative of iron production. Particularly high iron in copper artefacts and slag compositions low in silica (say below 15-20%) would be indicative of the formation though not necessarily the extraction of iron during smelting. Analyses of various slags with very low copper content have been published by Steinberg and Koucky (1974). The fact that no unambiguous iron smelting slags have been located in Cyprus favours the postulate that iron production was intimately linked with that of copper, and the two would have formed the same type of slag. Further research on empirical evidence of this kind is needed to substantiate or negate the model presented here.

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APPENDIX

NOTES ON THE THERMODYNAMICS OF SMELTING FURNACES

Definitions

Heats of formation and reaction (Table 5A) Almost all chemical reactions involve the emission or absorption of heat. When this applies to the formation of a compound from its elements it is known as the *heat (or enthalpy) of formation* of the compound (H). In a complex reaction, the algebraic sum of the heats of formation of all the reactants and products gives the *heat of reaction* (ΔH). The standard convention is that heat absorbed is positive while heat emitted is negative. This procedure permitted the calculation of the heat balances in the above Section **Fuel Shortage as the Trigger for the Iron Age**.

Free Energy of Formation and Reaction (Table 5B) A related quantity, also known as the *Gibbs Free Energy* (G) measures the tendency for a chemical reaction to take place. In such a reaction, when the algebraic sum of the individual free energies of formation (*Free energy of Reaction,* (**ΔG**) is negative there will be a tendency for the reaction to take place spontaneously. Where competing reactions are possible, the one which gives rise to the most negative free energy change will be preferred.

Entropy The third fundamental thermodynamic quantity, *Entropy* (S) is a quantity expressing the packing of the atoms in a system and is particularly sensitive to changes in volume of gases. It is related to enthalpy and free energy by the equation:

$$G = H - T\,S \quad \text{or} \quad \Delta G = \Delta H - T\Delta S$$

where T is the absolute temperature (°K).

Table 5A. Thermodynamic data: heats of formation. (K.cal. per mole at 298 deg. K).

COMPOUND	*HEAT OF FORMATION*
CO	-25.4
CO_2	-93.69
Cu_2O	-40.55
CuO	-37.74
FeO	-65.32
Fe_2O_3	-200.0
Fe_3O_4	-268.31
SO_2	-71.0
Cu_2S	-19.02
CuS	-12.7
FeS	-22.72
FeS_2	-42.52
$(Cu,Fe)S_2$	-42.52 (estimated)

Table 5B. Thermodynamic data: free energies for the reactions represented in the Ellingham diagram (Fig. 3). (K.cal. per gram atom of oxygen reacting).

Reaction	*Free Energy Change at specified temperatures*			
	298° K	*1000° K*	*1400° K*	*2000° K*
$C + ½O_2 = CO$	-32.81	-47.88	-56.21	-68.42
$½(C + O_2 = CO_2)$	-47.13	-47.26	-47.28	-47.25
$CO + ½O_2 = CO_2$	-61.44	-46.76		-26.08
$2CO = C + CO_2$	-28.63	+1.23	+17.89	+42.34
$2Cu + ½O_2 = Cu_2O$	-35.00	-23.19		-10.27
$Cu + ½O_2 = CuO$	-30.96	-16.20		+3.18
$Fe + ½O_2 = FeO$	-58.76	-47.85		-33.81

Though H does not change greatly with temperature, S can vary considerably where gases are involved, and as T is numerically large, temperature changes can have a large effect on G. Thus, a reaction which takes place at a lower temperature may reverse at a higher temperature if it should happen that (ΔG changes sign. In the reactions between carbon and oxygen for example, carbon dioxide has a higher free energy of formation below about 1000° K and is the oxide preferentially produced. Above that temperature carbon monoxide has the higher free Energy of formation and is the oxide formed.

Application

This is illustrated in 'Ellingham' type diagrams (Ellingham 1944, Richardson and Jeffes 1948, Charles 1980) in which free energy changes are plotted against temperature for related reactions. It is usual to plot the Free Energy vertically with the most negative value at the top. The one most relevant to the present work is that for reactions of oxygen with carbon and with some metals. Such a diagram is produced as Fig. 3, using Free Energies (per atom of oxygen reacting) calculated from data in the *Handbook of Physics and Chemistry* (1969).

The Ellingham diagram can be used to predict which of alternative possible reactions will take place, since there must always be a net decrease in Free Energy. Thus, a reaction which appears lower on the diagram corresponds to a more negative Free Energy change than one which appears above it and will therefore have precedence. Differing responses to temperature changes might cause the (ΔG versus temperature plots to cross, so that the priority of the reactions changes. In general, with competing reactions, the one which at a given temperature appears lower on the Ellingham diagram will prevail. The Ellingham diagram neatly illustrates the effect mentioned above for the C/CO_2 and C/CO reactions. Here the (negative) free energy of formation of carbon dioxide from carbon varies little with temperature, whilst that for carbon monoxide increases rapidly, exceeding the value for CO_2 at *c.* 1000°K (727°C). Above the cross-over temperature carbon dioxide will react with carbon to form carbon monoxide, while below that temperature, *in the absence of oxygen*, carbon monoxide can break down into carbon and carbon dioxide (see above, Equation 10).

The Ellingham diagram can also be used to predict the conditions under which metal oxides can be reduced by carbon or carbon monoxide. Thus, copper oxide can be reduced by either C or CO at any temperature above ambient, while the intersections of Fe/FeO line the with the (CO/CO_2) line (*c.* 800°K) and the (C/C0) and (C/CO_2) lines at 1000 and 1100°K indicate that reduction by CO is possible only below 800°K,

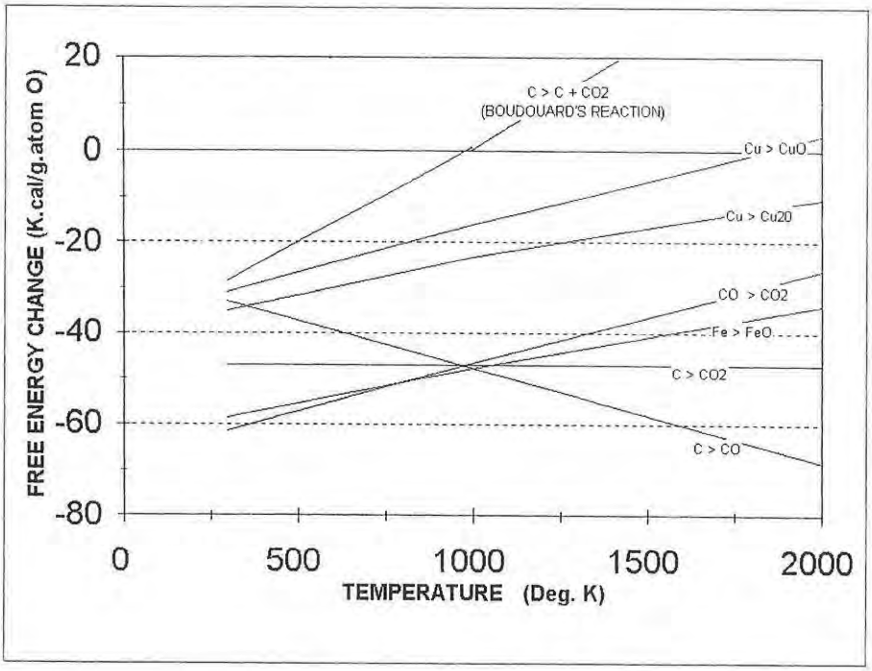

Fig. 3. Ellingham diagram for oxides.

and by carbon is possible only above 1000°K. Since reactions between solids are very much slower than those between solids and gases, the CO reaction accounts for 80-90% of the reduction of iron oxide to iron during smelting.

Table 6. Blind comparisons of analyses of standard bronzes.

Bronze standard	ELEMENT *Cu*	*Sn*	*Zn*	*Pb*	*Sb*	*As*	*Fe*	*Ni*	*Total*
UE13-1 [a]	83.90	13.70	0.62	0.22	0.29	0.23	0.45	0.11	
UE13-1 [b]	83.9	13.6	0.59	0.23	0.30	0.23	0.42	0.11	99.39
UE13-1 [c]	83.9	13.67	0.60	0.29	0.295	0.233	0.43	0.11	
BM183-3 [a]	84.5	6.69	3.25	3.40	0.25	0.15	0.028	1.52	
BM183-3 [b]	84.7	6.58	3.21	3.34	0.26	0.14	0.024	1.54	99.91
BM183-3 [c]	84.6	6.60	3.22	3.36	0.27	0.144	0.025	1.53	
BM c42-01 [b]	65.9	0.91	32.6	0.11	0.05	0.06	0.185	0.10	99.91
BM C42-01 [c]	66.0	0.82	32.6	0.12	0.05	0.06	0.185	0.10	

a = Analyses carried out by independent laboratories
b = Analyses carried out at the British Museum laboratories
c = Analyses carried out at Edinburgh (this work)

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