

This special MASCA Journal supplement is issued to coincide with the Science Fair which will be devoted to archaeometallurgy, at the annual meeting of the Archaeological Institute of America being held in Philadelphia, December 27th-30th. The supplement's articles themselves underline two facts. First, that archaeometallurgy has developed as a major element of MASCA's research activity over the past three years, to cover the metalworking technology of many cultures in a wide matrix of time and space. Second, that archaeometallurgy is also a rapidly growing discipline worldwide.

I am delighted therefore to take this opportunity to thank those outside the University of Pennsylvania for their tireless support of the MASCA program—specifically Michael Notis and Heidi Moyer at Lehigh University, Charles Swann at the University of Delaware, and Reed Knox (now retired from Alan Wood Steel)—and to express my appreciation for the excellent contributions provided by our colleagues abroad, contributions which make this supplement thematically well-rounded overall.

Stuart Fleming, Scientific Director, MASCA

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Cover: Carburized microstructure indicative of steel seen here as contiguous colonies of pearlite (grey, lamellar), and elongated, angular Widmanstätten ferrite plates were identified in a *circa* 12th century B.C. iron bracelet (I.55) from the Baq'ah Valley, Jordan. Structures as seen here are magnified 800 times.

THE EARLIEST STEEL FROM TRANSJORDAN

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Introduction

Previous issues of this journal (see McGovern 1979, 1981), described the discovery of an undisturbed burial cave (A4) in the Umm ad-Danānir region of the Baq'ah Valley in Jordan which dated to the earliest part of the Iron Age (*circa* 1200-1050 B.C.). Subsequent excavation of its unique burial deposits yielded the skeletal remains of 220 individuals and a full assemblage of associated grave goods. Among the most significant finds from these deposits are a group of eleven complete pieces of iron jewelry—eight anklets (or bracelets) and three rings—together with forty additional fragments. Three basic types of anklet/bracelet are represented: (i) with ends open, (ii) with ends overlapping and superimposed one above the other, and (iii) with ends overlapping side-by-side. Two types of rings are represented, each with a rectangular cross-section, but one with open ends, the other with overlapping ends. (See Plate 1 a and b for illustrations of these various forms.)

Waldbaum (1978) has published a survey of iron artifacts recovered from Eastern Mediterranean excavations. Five objects were listed for two Late Bronze Age II Palestinian contexts, together with about twenty more from various 12th century B.C. loci at seven Palestinian sites. The East Bank of the Jordan is represented only by a pair of bracelets and a pair of rings from a transitional Late Bronze/Early Iron Age tomb at Madaba (Harding 1953). These comprise one example each of an open-ended and an overlapping, superimposed type, as defined above.

To these must now be added an Early Iron Age knife from tomb 113 at Tell es-Sa'idiyeh (Pritchard 1980), an unidentified fragment from an LB II tomb on Jebel al-Nuzha near Amman (Dajani 1966) and part of an anklet/bracelet from an LB II burial cave (B3) in the Baq'ah Valley recovered in the summer of 1981. Thus, with the excavation of Cave A4, the number of iron artifacts from 12th century B.C. Palestine has been roughly tripled, while for Transjordan it has been increased sevenfold. This clearly represents a major statistical shift in the distribution of iron artifacts throughout the Eastern Mediterranean.

Techniques of analysis

A cross-section was cut from each artifact with a diamond impregnated cut-off wheel, then mounted in thermo-setting

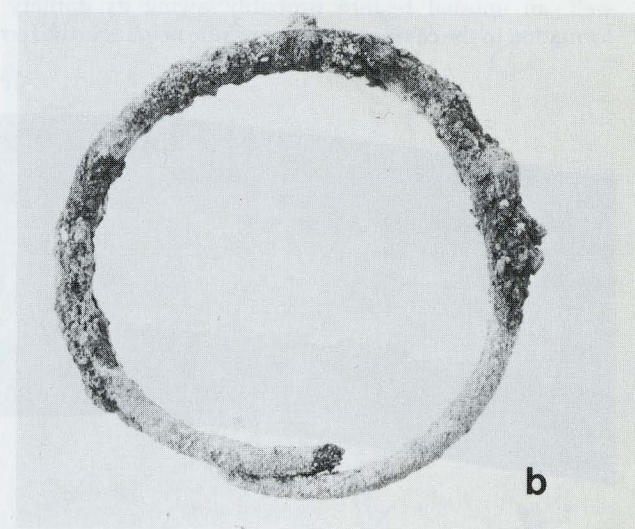
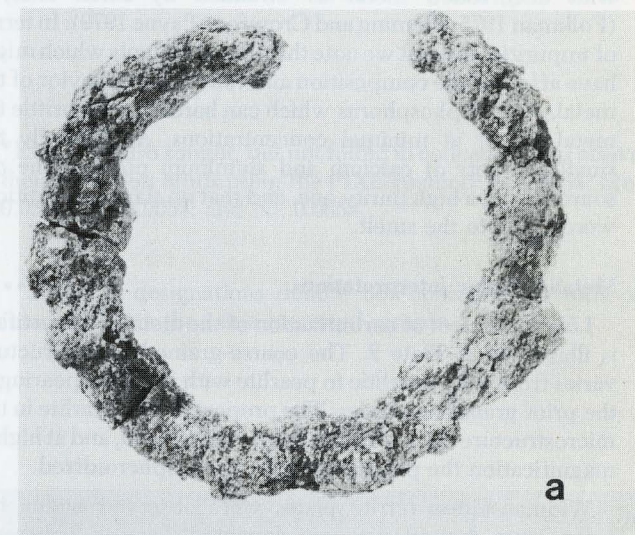


Plate 1:
Anklet/Bracelet types from Cave A4 (D., approx. 0.07 m, 0.06 m, respectively).
(Photographs: Courtesy of N. Hartmann, MASCA.)

plastic, polished on four abrasive papers and finally on three polishing wheels. All samples were studied in both the unetched and etched condition and photographed. The etchant used was 2% nital. When slag inclusions were found to be present, their chemistry was studied using energy dispersive x-ray analysis in a scanning electron microscope.

A preliminary analysis of five artifacts, the only specimens found to have macroscopic islands of uncorroded metal, is presented here. A total of seventeen artifacts in all were studied. The remaining twelve artifacts were found to be heavily oxidized and are now being studied for evidence of pseudomorphic ghost structures indicative of carburization (see Knox 1963).

Table 1 provides the elemental analyses of the five artifacts with uncorroded metal as obtained by PIXE analysis (Folkman 1975; Fleming and Crowfoot-Payne 1979). In terms of impurities present we note that those elements which might have affected the composition and functional behavior of the metal, such as phosphorus which can harden or embrittle the metal, occur at minimal concentrations. Additionally the small amounts of calcium and aluminum point to the ore source being a high purity one, and that no slagging additions were made to the smelt.

Metallographic interpretations

I.55: The extent of carburization of the metal of this artifact is illustrated in Plate 2. The coarse-grained microstructure varies from fully pearlitic to pearlite with ferrite appearing at the prior grain boundaries. The proportions of pearlite in the microstructure are consistent from field to field, and at higher magnification the pearlite appears to be spheroidized.

Widmanstätten ferrite plates were observed within the prior grain boundaries, as elongated angular structures running parallel to one another (Plate 2). However Widmanstätten patterning was also found within the grains as well, an unusual feature probably owing its stability or formation to the co-occurrence of nitride needles in the ferrite

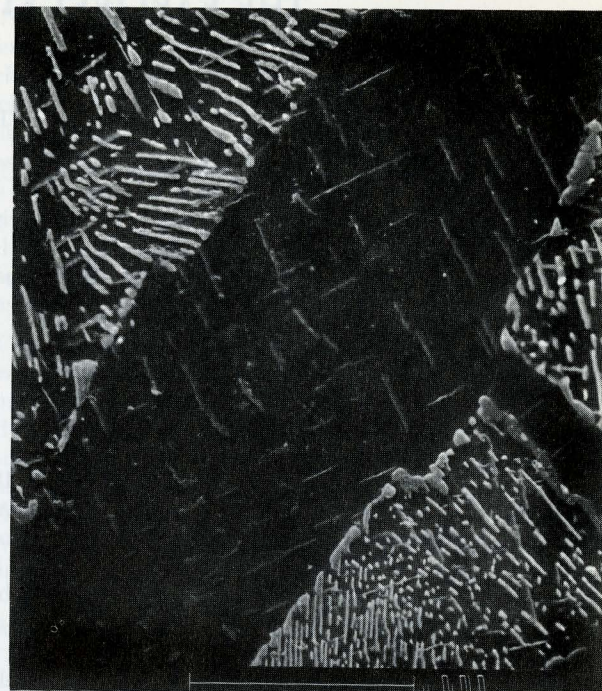


Plate 3:
I.55 SEM photomicrograph (3000X) showing nitride needles in ferrite surrounded by partially spheroidized pearlite.

phase (Plate 3). This structure is indicative of long-time exposure in the smelting furnace and/or the smith's forge, as well as of subsequent slow cooling. Additionally the SEM analysis of the fayalite (Fe_2SiO_4) slag-matrix of slag particles entrapped in the metal indicated that they contained a relatively high level of potassium, which would point to a charcoal fueled process, and only a small volume fraction of wüstite (FeO) indicative of the high efficiency and skill of the smelting operation.

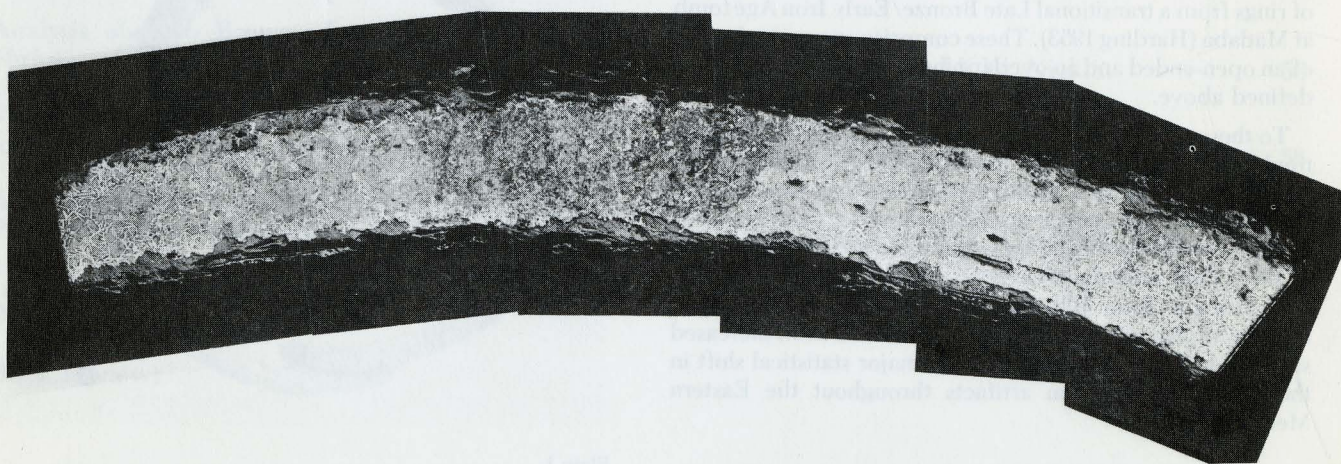


Plate 2:
I.55 Photomicrograph (24X) of 2 cm. longitudinal section of anklet/bracelet showing extensive carburization and Widmanstätten patterning.

Table 1

Elemental Composition (% by weight)*

Sample reference**	Fe	Al	Si	S	Ca	Co	As	Cu	Cr	Au
I.55	99.5	0.058	0.14	≤0.010	0.087	≤0.030	≤0.002	≤0.005	0.096	0.014
I.77a	99.8	0.042	0.051	≤0.010	0.039	≤0.030	≤0.002	≤0.005	≤0.010	≤0.010
I.77b	99.5	0.16	0.079	0.061	0.035	≤0.030	0.010	0.013	0.091	≤0.010
I.147	99.7	≤0.030	0.067	≤0.010	0.029	≤0.030	0.028	≤0.005	0.068	≤0.010
I.202	99.7	0.28	0.12	0.030	0.043	≤0.030	≤0.002	≤0.005	0.067	≤0.010
I.226a	98.9	0.10	0.22	≤0.010	0.12	0.37	0.006	0.007	0.073	0.015
I.226b	98.5	0.11	0.27	0.014	0.50	0.42	0.004	0.017	0.10	0.016

- I.55: Type b
Sample location: from terminal tip.
- I.77: Type indeterminable
Sample location: from broken edge of fragment.
- I.147: Type indeterminable
Sample location: from broken edge of fragment.
- I.202: Type b
Sample location: from terminal tip.
- I.226: Type a
Sample location: from terminal tip.

*Elements also sought, but not found in concentrations above their detection limits using the PIXE method: K, 0.063%; Mn, 0.050%; Zn, 0.006%; and Ni, 0.005%.

**a and b designations denote non-contiguous islands of intact metal in the same mounted sample.

I.77: One surface of this bracelet had a relatively high carbon content and a finer ferrite grain size, but the carbon content was still short of the eutectoid composition which would be about 0.7% carbon. The remainder of the cross-section revealed only minimal evidence of carburization distributed unevenly among large grains of ferrite.

Elongated slag stringers record the flattening which the metal underwent during forging (Plate 4). Working was extensive enough that certain stringers were themselves broken into smaller sets of aligned slag inclusions, with cracks running transverse to the direction of elongation. Clearly the forging was carried out at a lower temperature, *i.e.*, while the slag was brittle, not plastic.

SEM analysis of slag particles in this artifact indicated that they comprised a typical wüstite-fayalite composition with a relatively high volumetric contribution of wüstite. Also, only very low levels of sodium and potassium were found in the slag. These observations, when contrasted with the results for I.55 could indicate a highly variable smelting process, or a completely separate source of smelted iron.

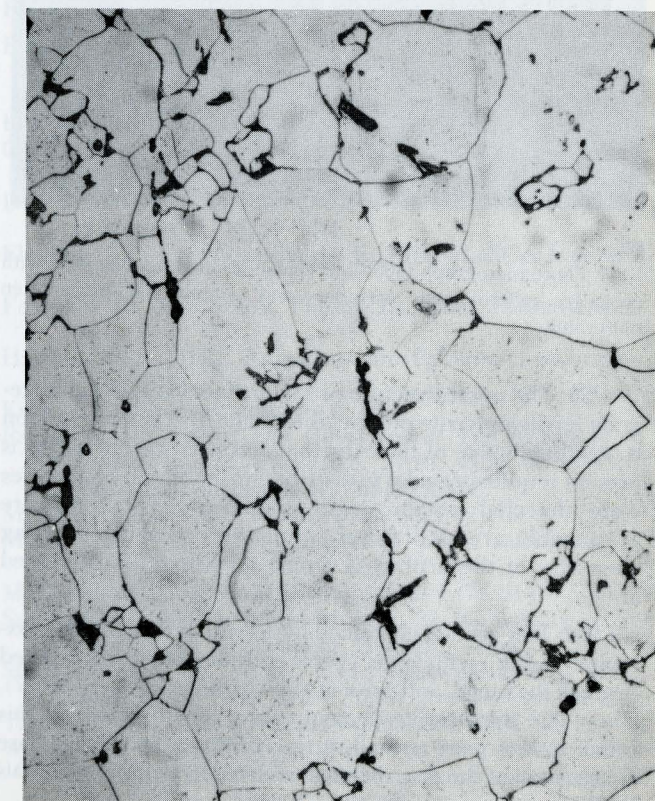


Plate 4:
I.77 Photomicrograph (400X) of zone of large ferrite grains showing minimal evidence of carburization.

I.147: The metal of this artifact is a coarse-grained pearlite steel (Plate 5), with a carbon content of about 0.7%, perhaps a little less because of the presence of a small amount of ferrite. There is evidence of partial spheroidization of the carbides, and Widmanstätten side plates had formed in hypoeutectoid regions along one edge of the bracelet. Slag inclusions appear to be mostly fayalite and of a single phase.

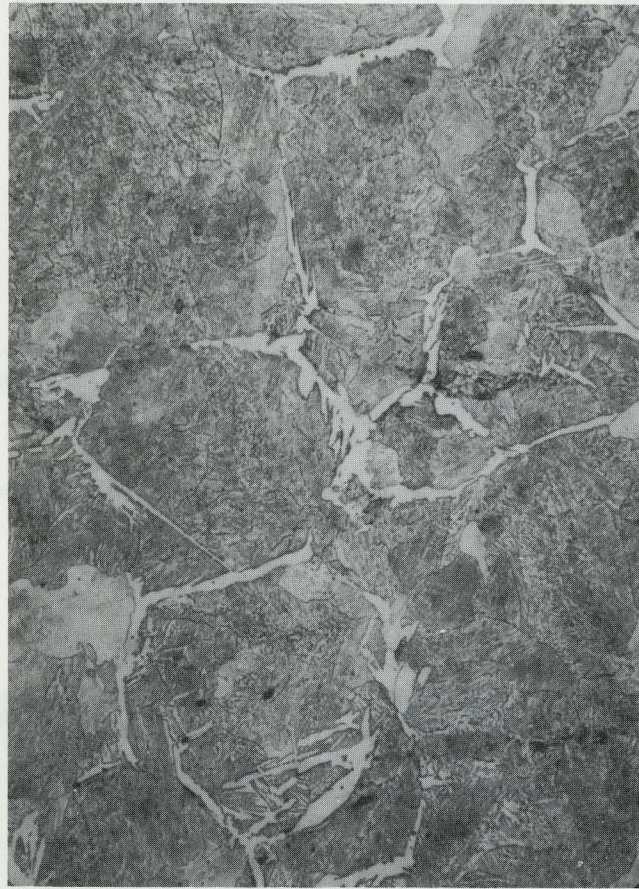


Plate 5:
I.147 Photomicrograph (400X) of coarse-grained pearlite steel with partially spheroidized carbides and angular Widmanstätten patterning.

I.226: The microstructure of this artifact varies from large-grain ferrite to ferrite plus pearlite (Plate 6). The composition is well below that of the eutectoid and what carburization is present is quite irregular in distribution. Subgrain boundaries were detected within single grains of ferrite, and are presumably a result of lower temperature working. The slag chemistry of this artifact is similar to that of I.77 discussed above.

I.202: This artifact contained hypereutectoid steel (approximately 0.8% C) with large, coarse carbides evenly distributed through the metal, and present even in the oxide (see Plate 7). They, like the pearlite present, were spheroidized. In this instance there were virtually no inclusions upon which to base a discussion of the slag chemistry related to production of this artifact.



Plate 6:
I.226 Photomicrograph (400X) of heterogeneous zone varying in composition from large grain ferrite to ferrite plus pearlite.

Preliminary conclusions

The initial results of this study of the five anklets/bracelets are both exciting and enigmatic. Four of the artifacts have a structure wherein there is a uniform distribution of carbon from surface to surface, making them the earliest verified instances of mild steel from Jordan and placing them with a small group of the earliest dated steel from Eastern Mediterranean sites such as Idalion on Cyprus (Tholander 1971) and Ta'anach in Palestine (Stech-Wheeler *et al.* 1981). Initial investigation of an additional twelve oxidized iron artifacts from Cave A4 suggests that the majority of these may also have been composed of a mild steel.

Why pieces of jewelry should be manufactured in steel is not clear, nor is the process by which this steel was produced clearly definable. The definitive evidence for solid state carburization would be the presence of a carbon gradient at or near the surface, *i.e.*, of carbon diffusion into the iron that would have proceeded from the exterior surface inwards. The metallography of the Baq'ah artifacts however, revealed no gradients of this kind in the cross-sections taken, in part because of surface corrosion, in part because the microstructure of the metal is so uniform. Thus it is not possible to draw a definite conclusion as to the origin of the material via carburization.

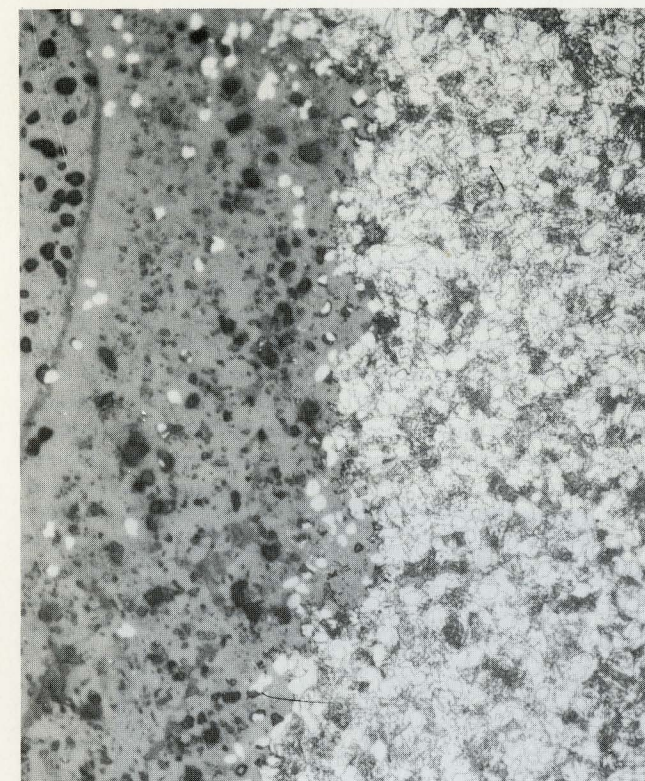


Plate 7:
I.202 Photomicrograph (400X) of large coarse spheroidized carbides evenly distributed in the hypereutectoid steel.

An alternative possibility can be suggested which would account for the microstructures evident in the Baq'ah steel. In most of the artifacts not only are the amounts of carbon near the eutectoid composition (0.7%) but also there are high concentrations of nitride needles present in the ferrite (Plate 3). This could be explained if the iron being reduced had reached a semi-molten (slushy) or molten state in the smelting furnace. Though the temperatures required to achieve this state, 1400-1475°C, are well above the normal range thought to have been routinely used in antiquity, they are not actually beyond the capacity of a bellows-blown furnace. Localized regions of the bloom, or the entire bloom itself, could therefore, under some unusual yet conceivable conditions have been made molten (see Smith 1966), yielding appropriately high diffusion rates and high solubilities of both nitrogen and carbon into the liquid iron.

However, though the Baq'ah steel artifacts show no indication of having been subjected to any heat treatment such as quenching or tempering, this metal would have had mechanical properties (*e.g.*, strength) which would have made it at least equivalent to the bronzes (with about 10% to 15% tin content) found in the same tomb.

In Cave A4, bronze artifacts were three times as prevalent as steel ones, yet intriguingly, none of these artifacts were weapons. The available archaeological and documentary evidence for the Late Bronze/Early Iron Age transition in Palestine suggests that it was far from peaceful. It is during

this period that the Israelites and Ammonites begin to consolidate their empires in the region.

We have no reason to believe the iron artifacts were imported. The proximity of iron ore deposits of limonite and hematite in the Wadi Zarqa and Ajlun regions, the nearest one being only 10 kilometers to the north (Basha 1968; Bender 1974), together with the fact that virtually nothing else recovered from Cave A4 apart from perforated Mediterranean and Red Sea mollusk species could be considered as imports, strongly suggests nearby production, perpetuating Bronze Age metalworking traditions.

Limited archaeological investigation in the Ajlun at the sites of Mugharat el Wardeh and Abu Thawab (Coughenour 1976) has thus far only been able to substantiate medieval Islamic smelting operations, but surface collection of sherds from the Roman/Byzantine and early Iron periods points to earlier activity. However, until these areas, including the Baq'ah, are more fully surveyed and well-dated metal-working installations and associated settlements found (*e.g.* Khirbet Umm ad-Danānir: see McGovern 1980), such a hypothesis, or alternative views which see the introduction of iron/steel in Transjordan as more abrupt and a result of an intrusive cultural element, remain unproven.

Acknowledgments

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