# SCIENCE

# **Man's First Encounters** With Metallurgy

Man's discovery of ores and metals helped to shape his sense of science, technology, and history.

# Theodore A. Wertime

Some 40 years ago the British Association for the Advancement of Science elected to investigate scientifically the composition and geographic origin of metals used in early artifacts. Similar investigations were gradually initiated in other countries. Today spectrographic analysis absorbs the talents of 15 or more organizations variously located between the east coast of the United States and the Caspian Sea. Thousands of copper artifacts have been sampled. No other science of the many now serving Near Eastern and European archeology can claim a comparable contribution to the study of its own origins (1).

Yet we are only beginning to understand the ramifications of man's first sophisticated uses of matter. Metals defined the technological and economic character of the era of urban life as surely as stone had defined the character of the millennia during which cavemen first tried to shape tools by hammering or cutting rocks they found on the ground or in the earth. A biblical passage alluding to the early iron age gives a glimpse of first ventures into the arcane world of ores and fire to be found underground (2):

and copper is smelted from the ore. Men put an end to darkness,

and search out to the farthest bound the ore in gloom and deep darkness.

4 DECEMBER, 1964

They	open	shafts	in	а	valley	away
fro	m whe	ere mei	n li	ve	;	
		-				-

they are forgotten by travelers, they hang afar from men, they

swing to and fro.

As for the earth, out of it comes bread; but underneath it is turned up as by fire.

During the first century A.D. metals were made the basis of a general scheme of history by two men of widely separated cultures: Lucretius, author of De Rerum Natura, and Yüan Kang, a writer of the Han dynasty. Each divided the history of mankind into three ages: stone (or bone), copper (or bronze), and iron. The ordering of history according to dominant material has become the most popular and useful form of chronological classification in our own day (3).

Whatever the place of metals in history, their various debuts as useful materials are not clearly defined or easily explainable historical events. The thousands of bits of evidence collected to date suggest that man first viewed metals as individual and puzzling representatives of a new form of matter. He began by hammering native gold and copper, or meteoric iron, but he did not appreciate their character until he had learned to melt and cast a variety of metals and, ultimately, to reduce them from their ores. Smelting was the crucial step through which he first gained an understanding of metals as the lustrous, ductile, malleable materials they are, and as derivatives, by strange processes of fire, of varicolored and manyfaceted mineral stones.

We surmise today that the discovery of smelting did not revolve merely about copper, the first industrial metal, but that it engaged man in chemistry that divulged, in relatively quick sequence, the existence of lead, silver, tin, and probably iron. Tin revealed itself as the ideal alloy in bronze only after long and often unintentional trials with impurities such as arsenic and antimony.

The assortment of ores confronting early man was staggering. Copper ores alone appear in more than seven varieties, evincing in color the yellow of chalcopyrite and the ruby of cuprite, the green of malachite, the bright blue of azurite, the purple of bornite, the purplish-brown of native copper, and the gray sparkle of chalcocite.

The birth of metallurgy must therefore be seen as the culmination of difficult and scientifically hazy labor, in the course of which men learned to extract a number of metals from their ores by fire and to cast and alloy them. The discovery of metals appears to have begun in the 6th millennium and to have been reasonably well advanced by 2000 B.C. It occurred through an area stretching from western and central Anatolia across the flanks of the Taurus and Zagros mountains to the edge of the central desert of Iran.

The time and place are not accidental. The post glacial epoch in southwestern Asia, particularly after about 8000 B.C., was a time of discovery and exploitation of the material world, during which tribesmen on the flanks of the Taurus, Zagros, and Alborz mountains became arbiters of biological evolution by taming animals and cultivating plants, and instigators of technological revolution by learning the uses of fire and the potential uses of earth (4).

Archeologists cannot agree on the precise juxtaposition of mountain and

Iron is taken out of the earth,

Mr. Wertime is the editor of Forum for the Voice of America and was formerly U.S. cultural attaché in Tehran. He is the author of *The Com-*ing of the Age of Steel (University of Chicago Press, Chicago, 1962).

plain at which the pastoral arts of Abel, the agricultural arts of Cain, and the metallurgical arts of Tubal Cain were each learned. But it is doubtful today that the earth that yielded Cain's bread was also that which was "turned up as by fire." With some notable exceptions, the rich soils of the Mediterranean littoral or the river valleys of the Fertile Crescent, where agriculture reached its first apogee, are nearly barren of minerals and indeed poor in fuels.

The conditions for the emergence of metallurgy were probably best fulfilled in the semiarid mineralized zones of the Anatolian and Iranian plateaus (5). It was possibly an unconventional ancestor of the tribal gypsy, dwelling in "parched places in the wilderness," accustomed to trading over vast distances, unafraid of the challenge of searching out ore in gloom and deep darkness or of the magic of its transmutation into useful tools, who presided at the birth of metallurgy. Such men still roamed the Iranian desert as late at twenty years ago, smelting their copper in basins in the sand.

But whoever these discoverers were, and however untamed, they were propelled by certain ineluctable social and ecological circumstances: The settled agricultural communities which came into being with the inception, ten thousand years ago, of the food-producing revolution, needed more versatile goods and media of exchange, and surplus human energies were available to fabricate these.

Man's exploitation of his environment had acquainted him with a wide variety of stones, minerals, and fuels.

In fire-using industries such as the making of pottery and bricks and the glazing of ornamental objects, artisans had achieved temperatures well above the melting point of lead and close to that of copper and had become acquainted with the alchemy of fire.

A favorable juxtaposition of mineral and fuel resources was to be found at sites in the uplands of the Middle East. Contrary to the belief expressed by many, the heavily forested zones of the Caspian would not have favored experimentation with pyrometallurgy. But in the semiarid regions of central Anatolia and Iran, outcrops of useful rock and ore present themselves in vivid hue. Here obsidian and, if the Afghan extension of the Iranian plateau is included, lapis were first exploited. These zones were once (and to some extent still are) the home of significant stands of two plants, wild pistachio and Haloxylon amodendron (known colloquially as "tagh"), which provide metallurgical charcoal of superlative quality. Recent pollen studies suggest that the flanks of the Zagros were forested with wild pistachio during a period that extended from 10,000 to 5,500 years ago (6).

Table 1. Progress in metal working (4500-2000 B.C.) (redrawn by permission of Cornelius Hillen, Rotterdam, Netherlands).

Year (B.C.) IRAN		MESOPOTAMIA		SYRIA	EGYPT	TURKEY
	South	North	(Amouq)			
2000 -		Isin-Larsa-				
		Urill		1	First Interm	Early Bronze e
		Akkodian				
	Hissor III	Early Dynastic	Ninevite	н	Old Kingdom	Chalco- lithic
7000			•			Proto-
3000-	11:20	Proto-		G		Chalco-
Hisso	Hissar 	Literate	Gawra	F	Gerzean	lithic
		Warka		 E	+	
	Hissar		+	<b>E</b> 77777777	Amration	Mersin XVI
		Ubaid	Ubaid	D		
4000-	Chesmeh	(Eridu)				(Beyce-
	All	1	Halat	B	Badarian	sultan)
45.00	Siyalk		Hassunah	X // //	Tosian	
4500 -		9	¥ <i>\\\\\\\\\\\\\\\\\\\\\\\\\\\\\\\\\\\\</i>		X/////////////////////////////////////	(Çatal Hüyük)

Hammering and annealing of native copper. Melting and open mold casting of native copper. Smelting and closed mold casting of copper from ores. Bronze (★)

Terrestrial iron, isolated occurrences.

First Appearance of Metal Objects

Among the oldest metallic objects that have been found are copper tubes decorating the ends of string skirts found by Mellaart in 1962 at Çatal Hüyük on the Konya plain in Turkey (7). The artifacts of Çatal Hüyük testify to a great surge in technology near the beginning of the 6th millennium B.C. The discovery of copper artifacts, which in some cases show technical and artistic transitions from woven baskets, next to early specimens of pottery and textiles suggests that here as elsewhere archeology is near the very threshold of the age of metals (8). These tiny copper samples have been provisionally identified as being of hammered native copper (9). Historians of technology eagerly await the further elucidation of this discovery.

Çatal, the dates of which are still very much at issue, merely illustrates once again how troublesome is the relative chronology of metallurgical finds. A fair portion of archeological dating of metals has been circular; the designation of a "chalcolithic" stage that is supposed to overlap the neolithic period is still largely based on the happenstance assignment of discovered artifacts to this stage. Moreover, the results of spectrographic analysis, despite the widespread use of this method,

1258

are often least available where most needed for dating the earliest artifacts. Nonarcheologists must step very gingerly in this area, which is still much debated among archeologists.

During the past decade the chief finds relating to the beginnings of metallurgy have been made in Anatolia. Previously, attention had been focused largely (though not exclusively) on Iran, northern Iraq, and Syria. The "pre-Anatolian" data are summarized in Table 1. This chart puts metallurgy into the conventional archeological sequence of pottery evolution and may be compared with Table 2, which attempts to relate all three of the major pyrotechnic arts—pottery, metallurgy, and glazing and glass-making—on a single time scale.

From the data from Anatolia and from a study of their relationship to earlier discoveries of ancient metal objects, David Stronach concludes (10)

that the art of the smith was fairly well developed and widespread by late in the 5th millennium B.C., a conclusion which is reinforced by the Çatal finds and more recently by other Anatolian finds. Table 3 gives a highly selective list of objects found in sites whose dates are reasonably well established and shows the pattern of developing metallurgy. This list cannot possibly include all sites of importance to the beginnings of metallurgy, particularly those of somewhat later vintage. Nor can we in this abbreviated treatment deal individually with those many spectrographic analyses that suggest statistically how man progressed to using the more impure and chemically complex ores, learning about matter in the process.

The earliest hoards of metal objects so far discovered date largely from the beginning or middle of the 5th millennium, if the antiquity of artifacts of Tepe Sialk in Iran, which is still arguable, is accepted. These metal finds reveal that by the 5th millennium copper, lead, silver, and gold were known at various sites in the Middle East (11-15).

It is impossible in a discussion as brief as this to examine all the metallic forms among the oldest items archeologists have discovered. A copper bead found in level 12 of Chagar Bazar and some copper balls uncovered in substratum 3 of period I at Sialk appear to be of the most primitive type, and mostly on the order of the finds at Çatal Hüyük. The bead at Chagar was found to be made of almost completely pure copper and showed no traces of arsenic, nickel, tin, zinc, or sulfur. Such purity suggests that it, too, was of native copper. Regrettably, it was not submitted to metallographic analysis to determine whether it had been merely hammered cold or had also been subjected to some degree of heat.

Pottery	Metals	Glazes, paints, and glass	
	Before 6000 B.C.		
	· · · · · · · · · · · · · · · · · · ·	Ochre used in funerary practices; cave paint- ers employ crushed oxides. Finds of rouge and eye shadow show crush- ing of hematite, galena, malachite.	
Çatal Hüyük pottery shows phases of pottery evolution.	6000-5500 B.C. Copper tubes, Çatal Hüyük.		
First Jarmo pottery. Sialk red wares use iron oxide slip.	5000-4500 B.C. First metal hoards in Anatolia and Iran suggest hammered-annealed copper.		
	4000 B.C.		
Closed kilns at Sialk III.	Cast Halaf and Sialk objects indicate melting of	Badarian glazes in Egypt bring together	
Ialaf polychrome wares follow metallic forms.	copper, smelting of lead, silver, and possibly copper. Extensive metal finds in Iran. Badarian Egypt shows first evidence of copper.	crushed ore and alkali in closed chamb at temperatures not exceeding 850°C.	
Ubaid wares show achievement of higher temperatures, first reducing conditions.	3500 B.C. Widespread casting of copper on Near Eastern plateau involves many impurities, particularly arsenic, lead, nickel, and tin. Meteoric iron ob- jects found at Gerzeh. New metallurgy gradually spreads to Egypt, along with other influences from Mesopotamia.	Slags begin to suggest nature of glazes. Egyptian blue comes into vogue.	
	3000 B.C.		
Potter's wheel comes into use at Amouq and elsewhere.	Age of metals begins great flourishing expansion in Mesopotamia as bronze first tentatively ap- pears, then gradually asserts industrial strength. Lead bronze castings of Uruk give way to purer tin bronzes of Ur. Similar developments in Syria, Azerbaijan, and elsewhere. Silver is medium of exchange, gold appears in statuary and jewelry. Bronze and lead dominate castings. Metal tools for cutting, digging, and shaping are common by Jemdet Nasr phase.	Glass beads first appear in Egypt, soon appear also in Mesopotamia.	
	2500 B.C.		
		Glassmaking, employing metallic colors, be gins to flourish in Egypt.	
	2000 B.C. Trade in metals is widespread in mid-East. Bellows depicted ca. 1500.		

4 DECEMBER 1964

Not surprisingly, however, the earliest smiths began at once to reproduce in metal the objects which once had been fashioned of stone, bone, or wood. The first objects to be found at Sialk (period I, substrata 3 and 4) are a hammered round awl, two pins with biconical heads, a needle with a forged eye, and a spiral. These forms had been developed by period II into tanged awls, a button, and a bracelet, and by period III into large copper pins, punches, and cast axes. Two silver buttons were found from period III.

At Arpachiyah, the site in northern Iraq which is richest in Halaf polychrome ware, a piece of lead 0.042 m high and 0.12 m in diameter, as well as fragments of copper pins, was found. A later cast chisel of probable Ubaid date has also been uncovered.

Mersin and Beycesultan, two Anatolian sites dating from the 5th millennium, round out the catalog of very early artifacts; I shall consider the first objects made of terrestrial iron in their subsidiary place in the history of smelting.

Nail-headed and scroll-headed pins turn up in level XXI of Mersin, succeeded by a copper chisel in level XVII, and pins, chisels, and axes at level XVI. At Beycesultan, a 5th-millennium storage jar yielded no pins, but did contain a silver ring, copper needles, several chisels, awls, and a fragment of a dagger blade.

One may infer not only that a relatively wide variety of metals and metal types appeared in the interval between Sialk I and III, but also that a wide variety of techniques must have been employed. The first copper objects from this millennium and a half were presumably hammered, but the art of casting in molds made an early appearance at Sialk and possibly Arpachiyah and Beycesultan. As I shall suggest, hammering does not rule out the probability that the copper was annealed or even purified by melting. The existence of lead objects raises the speculation that smelting of ores was already known, but in any event it is a clue to the artisan's growing sophistication about the various metals (16).

In this regard, spectrographic analyses of two of the earliest samples of Sialk artifacts are instructive (17). Two copper objects, a piece of a pin evidently from period I or II (18), and part of an arrowhead (19) were analysed spectrographically and metallographically (Table 4). From their rela-

Table 3. A sample of earliest metal finds.

Site	Approximate dates (B.C.)	Objects	
Sialk I-III (Iran)	4500-4000	Copper objects, silver buttons	
Arpachiyah (Iraq)	Late 5th millennium	Piece of lead, copper objects	
Chagar Bazar (Iraq)	Late 5th millennium	Copper bead	
Mersin (Anatolia)	Late 5th millennium	Copper objects	
Beycesultan (Anatolia)	Late 5th millennium	Silver ring, copper objects	
Chagar Bazar	ca. 3000	Lump of terrestrial iron	
Tell Asmar (Iran)	ca. 2700	Sword or dagger blade of terrestrial iron	

tive purity, the similarity between the proportions of minor contaminants in each, and their inner structures, it appears that both items embodied a form of the native copper common to the Kashan area. It is doubtful, though not impossible, that coppers from malachite or cuprite ores could have been refined to this degree of purity.

The Sialk pin had evidently been worked, annealed, and extensively reworked. The later arrowhead had definitely been remelted and cast (though few if any impurities other than oxygen were picked up in the process), and finally annealed. It appears from these studies of two evidently early samples from Sialk that at this strategic site the metallurgical art had progressed from the earliest techniques to a stage beyond the first primitive cold hammering of native copper.

#### **Probable Evolutionary Stages**

An order in man's first efforts to treat metals is thus discernible from the tangle of technical evidence (and lack of evidence) (20, 21). The view that pyrometallurgy was somehow confined neatly to the various phases of working copper until bronze and later iron came into their own is questionable, for the artifactual evidence may be highly misleading: Iron and silver all too frequently have corroded away or have been overlooked in their oxide or chloride forms, and lead is thought of as a worthless material (see Table 5).

It is possible to distinguish four stages in the development of man's treatment of natural copper:

1) Hammering. The earliest use of natural copper involved an extremely limited Stone Age technique by which small, specially selected pieces of metal were made into beads and possibly awls, pins, or hoops through cold forging. Attempts to cold hammer small pieces of native copper, described by Coghlan (1), show varying degrees of success. On the basis of recent experi-

ments by Cyril Smith, who succeeded in cold hammering pieces of Iranian native copper into small useful shapes, it seems that cold working offered slightly greater possibilities than sometimes believed. Native copper appears at Çatal, Sialk, and Chagar, but its presence seems doubtful at the earliest Egyptian sites, which date from a later time.

2) Annealing. The potentialities of the stoneworking techniques of simple shaping greatly increased when the smith learned to render copper into more malleable forms by annealing, a process involving softening in gentle heat. This was a step toward the melting of copper.

3) *Puddle Casting.* When the artisan discovered that melted pieces of copper formed a single puddle that would reharden, he had found a way of using up his scrap, thus enlarging his effective supply of metal.

4) Open Mold Casting. The first efforts to shape the molten metal that gathered in crevices in the hearth con-

Table 4. Composition of two Sialk artifacts and of native copper found at Talmessi.\* Code: a, 50%; b, .1-1%; c, .01-.1%; d, .001-. .01%; e, .0005-.001%; f, .0001-.0005%; g, < .0001%; ND, not detected. No elements were present in amounts from 1 to 50%.

Metal	Arrowhead (Sialk artifact 1425)†	Pin (Sialk artifact 1281)†	Talmessi native copper
Cu	a	a	а
As	ND	ND	ND
Sb	с	ND	ND
Sn	ND	ND	ND
Pb	<i>d</i>	ND	ND
Ag	d	d	b+
Ni	d-	ND	ND
Fe	d	d	d
Со	ND	ND	ND
Tl	ND	ND	е
Si	<i>d</i>	d	d
Мо	ND	ND	ND
Bi	<i>d</i>	g	g
Mg	е	e	d
Mn	ND	ND	f
Ca	d-	<i>d</i>	С
Al	ND	е	f
Zn	ND	ND	С

\* Determined through spectrography by the American Smelting and Refining Company. † Ghirshman's numbering. sisted in cutting forms in slabs of stone or of molding forms in clay. To fill these molds, the smith transferred the molten metal to a crucible, which might be merely a new bottom for the hearth. He was thus able to shape objects readily and to save the time required for hammering and annealing.

## **Smelting of Ores**

The early artisan thus almost simultaneously was following two important courses of discovery, one having to do with the shaping and, eventually, the melting of native metals, the other involving the application of heat to a wide variety of ores, clays, and other materials. I shall say more later about the chemical accompaniments of heat as applied to matter. Evidence of early casting of copper is sufficiently widespread to indicate that by the 5th and 4th millennia artisans in the great plateau were able to achieve temperatures high enough to melt copper, as well as to reduce all the major metals, including iron.

One can only speculate that, with the advent of smelting, feverish experimentation with ores, woods, and charcoals, and with furnace design, blowing devices, and clays, was carried out. Excavations of a copper refining site at Amouq, Syria (late 4th millennium), stir surmise that cuprite and malachite were jointly reduced in the presence of charcoal (14). Primitive practice at Ergani in Turkey, Anarak in Iran, or Wadi Arabah in Palestine suggests that sulfide ores were at some point mixed with these. From a study of pottery kilns from Sialk, domed brick furnaces from Mesopotamia, primeval assay and smelting furnaces from Iran. and 2nd-millennium copper smelteries still standing at Wadi Arabah, one can form rough notions of the evolution of furnace design, which need not have been well advanced to carry out the smelting of copper or lead. With proper blowing devices (which presumably appeared much earlier than inscriptions show) a simple hearth will do (22). Very hard scrub woods were available throughout the plateau. Charred pistachio is an excellent metallurgical reducing agent, and "tagh" in its uncharred, dried form has excellent thermal qualities and leaves little ash.

At some unknown point during experimentation with the various strata of ores, smiths discovered that sulfide

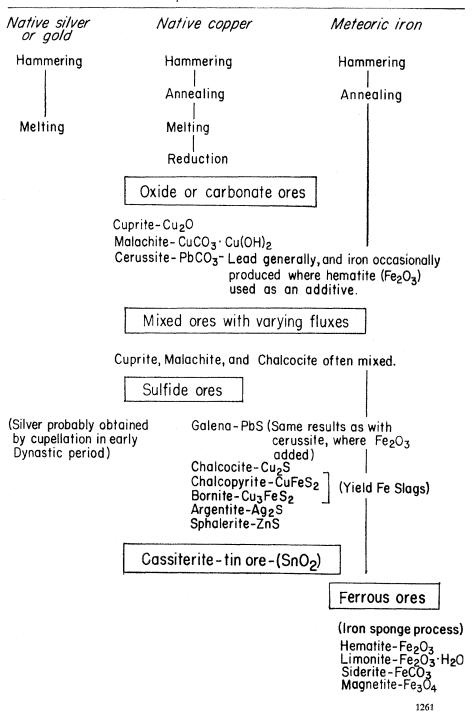
4 DECEMBER 1964

ores contain copper (or lead or silver or zinc) and thus began the derivation of these metals from the low-grade ores which are still used for this purpose. As experimentation continued, these smiths evolved techniques of roasting and, later, of matting and fire refining which enabled them to eliminate both sulfur and iron from the ore.

Smelting thus came to involve highly complex chemistry. F. C. Thompson believes that the impure "bronzes" which appeared in the 4th millennium, after the initial period of native copper, were produced largely as a result of smelting of sulfide ores; thus arsenic, antimony, and other similar elements were introduced into the metal (21).

This is a tempting theory, in view of the half-refined or black sulfide forms of copper delivered from the Persian Gulf, Ergani, and King Solomon's mines in the 3rd millennium, and especially in view of the large number of impurities found in copper artifacts of the 4th millenium (22, pp. 85-6; 23,





24). A large number of nonstannic impurities continued to be present in bronze until about 3000 B.C., when tin was found to be a manifestly superior alloy. The gradual diminution of such impurities after this date indicates that smelting (rather than casting) practice had improved.

At the beginning of the 3rd millennium, while artisans were still sorting out the elements uncovered by the fire of the smelting hearth, the art of writing was being evolved by Sumerian priests (22, p. 30ff.). The Sumerians and Akkadians had distinct words for gold, silver, and copper, although how the various forms were distinguished is uncertain. The Sumerian word sudogan has no exact translation, but it suggests zinc, arsenic, antimony, or possibly even lead. Tin came to share with lead the generic Akkadian name anāku (Sumerian ku-an·na) until its distinctive Indic name, nagga, emerged some centuries later. Because tin ores were quite scarce and because the metal served mainly as a component of alloys, tin as a reduced metal continued to be a subject of mystery and confusion.

Lead, which on the basis of remaining artifacts as well as of lexicography appears to have been discovered much earlier than tin, was likewise chiefly associated with other metals. The word a-bar (a-gar) seems at the time of Ur III (late 3rd millennium) to have meant a low, vulgar metal with many of the attributes of lead or occasionally those of tin. The use of lead in artifacts had become more common since the 5th and 4th millennia, when lead objects were first produced in Arpachiyah, Amouq, and Anau (12, pp. 12-17, 119). Lead was used in statuary at Uruk and Ur (both in its pure form and in copper alloys), in tumblers at Jemdet Nasr, in goblets at Tepe Hissar in northeastern Iran, and in human and animal figurines and sinkers for fishnets in predynastic and dynastic Egypt.

Because lead ore can be reduced at a lower temperature than copper ore, lead may actually have been smelted earlier than copper. But the most important role of lead in smelting may have been its usefulness for separating iron in a useful and recognizable form from its ores. Smelted iron was first found in a lump in level V at Chagar Bazar, dated provisionally between 3000 and 2700 B.C. That it soon found a limited industrial use is suggested by a fragment of a dagger or sword blade

found by Frankfort in a contemporary stratum (around 2700 B.C.) at Tell Asmar, which lies in alluvial Mesopotamia near modern Baghdad (15).

Iron was not mentioned in economic records of the first Sumerian and Akkadian cities, but it was noted in Mari, an 18th century B.C. Babylonian city on the Euphrates. There it appeared as the stuff of jewelry, very high in value. Iron fragments have turned up in archeological finds at Mari, confirming the literary record. Iron and gold rings are mentioned in a text of Susa of the same period (12, p. 119ff.; 25, 26), and iron arrowheads in the tablets of Alalah; in Babylon iron continued to be worth eight times as much as silver.

The Akkadian name parzillu, for iron, seems to involve the element bar or par, of uncertain meaning. Although there is probably no relationship between the logographic writings an-bar for iron and a-bár for lead, there is little question that the smelting of lead was an important overture to the recognition of iron as a useful, reducible metal. In ancient mining areas near Yazd, Iran, Cyril Smith and I examined a "bear" of iron, 4 feet in diameter, issuing from a semimodernized lead smelter. Such bears were common in the early days of lead smelting in Nevada.

In traditional Iranian practice, lead ores are reduced by a process known as the iron ore flux process, in which iron oxide is added to the charge. With an excess of charcoal and a heavy blast, giving hot, highly reducing conditions, the iron will be reduced; since it has a slight temperature-dependent solubility in the lead, the iron ore will eventually collect as an infusible lump of fairly compact iron known as a "bear." In time, experiments with this new metal, iron, would indicate that it could be attained in industrially usable form only by carefully controlled smelting temperatures, which keep the metal in a spongy state. But at least it was identified, and the interaction of elements in fluxing, which is so important in the reduction of sulfide ores carried out today, was recognized.

As metals were reduced from ores, impurities were also reduced, and from these the coppersmith learned of the hardening property of alloying elements. By 3000 B.C. he had identified tin, in proportions of one in ten, as the ideal agent for improving the casting properties and hardness of copper. The improvements brought about in the fusibility of copper by alloying it were accompanied by various discoveries concerning the value of a twopiece mold of stone or fired clay and, ultimately, of single-piece molds made about a wax model, known as the lost wax (*cire perdue*) process.

## Sites of Metallurgical Discovery

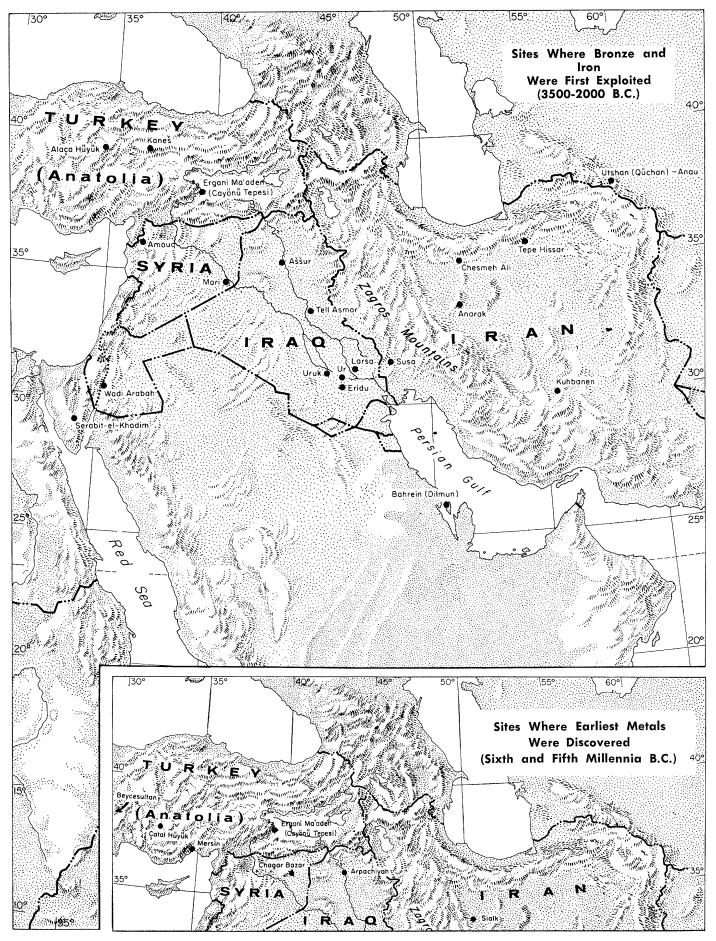
After 40 or more years of concentrated research in various fields, we know a great deal more about the four millennia of metallurgical evolution which culminated in the Mesopotamian qurqurru, the first metalworker to be given a professional name (22, p. 15).

Unfortunately, all the sciences brought to bear on metallurgical history have not yet shown precisely at what mining or smelting sites the important steps were taken. Archeology has not made a last concerted effort toward a scientific breakthrough, which would entail thorough scrutiny of old mines, slag heaps, and artifacts hitherto unanalysed. Current methods of analysis, moreover, have not been sufficiently refined to detect the very minute trace elements which might indicate more precisely the mother ores from which an artifact is made. Lacking the sort of inscriptions that identify historically the slag heaps of the Sinai peninsula, the archeological metallurgist has wandered about in "gloom and deep darkness," trying first nickel and then other impurities for the key to the actual sources of the copper ore used by the first artisans of the Middle East.

Two ancient centers of copper mining stand in reasonable propinquity to the metal-yielding archeological sites that stretch from Beycesultan in western Anatolia to Sialk in central Iran. They are Ergani Ma'aden in central Anatolia, and Anarak-Nachlak, about 80 miles from Sialk (27).

Other possible sources of native copper for the communities in Anatolia were deposits stretching eastward from Ankara, one in Corum province, one in Tokat, and one in Gümüshane. One such native copper analysed at Ankara was found to be 99.83 percent pure (28, 29).

Ergani once had native copper of reasonable (97.08 percent) purity, but presumably this supply was largely exhausted by the Old Babylonian period, ca. 2000 B.C. (23, pp. 337-41; 30).



4 DECEMBER 1964

On the basis of still-unpublished discoveries made by the University of Istanbul-University of Chicago expedition, Ergani may prove to have been the source of native copper for the earliest metallurgists. We know from slag heaps and economic records that this still-rich deposit was extensively exploited for exports to Assyria during the 20th and 19th centuries B.C. Smelting of its rich sulfide mineral, from which a black copper was initially extracted, must have begun there at an earlier date. This was primitive smelting indeed, to judge from the copper residue of nearly four percent which is found in the earliest slags. Does this represent the first attempt at reduction of sulfide ores? Possibly so.

Anarak and its environs yield appreciable quantities of ores of copper, lead, zinc, and nickel. Slag dumps abound, but the only documentary record consists of an inscription found 2 meters below the surface in a mine at Nachlak and not yet deciphered. However, the Talmessi mine at Anarak still affords native copper in quantity and of a composition (as revealed by spectrographic test) which suggests it might be the substance of the artifacts at Sialk (see 31 and 32).

At Anarak oxide and carbonate copper ores, though few, supplement the numerous sulfide ores. Until 20 years ago, the traditional method of smelting was still employed by native metalworkers, who mined a mixture of oxide and sulfide coppers. After applying a primitive pounding technique, they roasted the copper mildly. The product was reduced in small 6-foot furnaces, and copper of 97 to 98 percent purity was derived. Iranian practice thus seems to have frozen in a stage transitional between the smelting of oxide ores and the smelting of sulfide ores.

At Anarak nickel and lead ores are found in close juxtaposition to the copper ores. Since nickel and lead were prominent contaminants in various samples of Middle Eastern artifacts of the experimental period of the 4th and early 3rd millennia, further scrutiny of the Iranian Kavir as a very early source of supply for copper (and lead) is in order. Only in the early 20th century were the magnificent desert fuels of the area exhausted. Copper and lead slag heaps tell a story of millennia of exploitation, very possibly related to the foundation of the three sites, Sialk, Tepe Hissar, and Chesmeh Ali, all

By the late 3rd millennium, copper was being imported into Mesopotamia from Magan and Meluhha, whose location may have been on the Persian Gulf or on the Red Sea. As trade expanded in the days of the Larsa Kings, deposits in Dilmun (Telmun) on the Persian Gulf (Bahrein) were exploited. At the same time, Ergani copper was finding its way south to Assur in the north of Babylon (28, 33).

# The First Use of Tin

The question of man's first sources of copper is murky indeed. But the early use of tin, which is geologically very scarce throughout the Middle Eastern plateau (31, 34), is an even greater mystery. Most deposits which have been considered possible sources of tin in Iran and Turkey do not in fact contain tin-bearing ores. Utshan-Mian-Abot (Qüchan), near Meshed, is situated in a region of Iran (Khorassan), mentioned by Strabo as a source of tin (22, p. 85-6; 33, 35). But modern geologists who have visited the site have discovered no traces of tin-bearing ores. Kuhbanan, a remote city of the Iranian central desert, famous for its zinc oxide since Marco Polo's visit, has been similarly mentioned, but without geologic or historic proof.

In the broad zone formed by Turkey, the Caucasus, and Iran, geologists find only limited possibilities of the existence of tin ores in western Turkey. Azerbaijan, and the Caucasus (36). Ancient trading records from the days of Ur III onward attest to a substantial trade in tin from the borders of Iran, if one can overlook the uncertainty of the meaning of the word anaku. Mari, an 18th-century city of the Euphrates, was an entrepot for significant amounts of anaku derived from Elam and intended in part for westward movement, in part to remain in Mari's own depots for copper (25, p. 293; 33, 37). Anāku in large amounts is mentioned in a similar context in documents discovered at Tell Shemshara, near Iraqi Kurdistan. In Old Babylonian times, tin was transported, again in large quantities, from Assur on the Tigris to Kanes in Anatolia. The juxtaposition of evidence pointing to Iran as a source of tin suggests that geologists should

look more seriously at the assertions of Hamd-Allah Mustafi, a Persian writer of about A.D. 1340, who referred to a significant tin deposit on the Man-Rud River in lesser Lurestan. Here, he said (in a passage reminiscent of Pliny's and Agricola's references to alluvial mining) pieces of tin were to be found in large acorn-like shapes (38).

# Metallurgy and Pyrotechny

The smith of the late 4th millennium B.C. must have been remarkably sophisticated in a practical way regarding the individual phenomena of metallurgy. He must have known the effects on metals of hammering, annealing, oxidation, melting, and alloying; he must have been aware of the phenomena of simple decomposition of ores, their reduction, double decomposition, and metathesis (exchange of impurities); and he undoubtedly knew something of the miscibility and immiscibility of solutions. To follow his trail into these arcane revelations is indeed an exciting, if occasionally bewildering, quest.

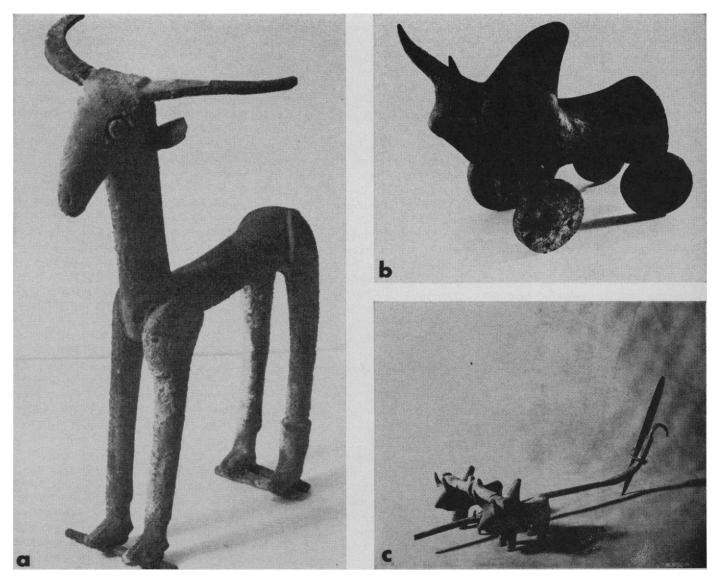
The study of the early history of metallurgy in the 20th century has been hampered by a rather single-minded attention to firing temperatures as the key to the appearance of the metals. Thus by *reductio ad absurdum* the Copper Age precedes the Iron Age by some 4000 or 5000 years because copper melts at only 1083°C, whereas the melting point of iron is 1537°C. But early smiths viewed not one element at a single temperature, but the whole world of matter on an ascending scale of heat.

One can imagine the bafflement of the first men to attempt smelting only by reviewing some facets of chemistry which would have seemed to them utterly contradictory. At 100°C oxide films first appear on some metals. At 330°C silver oxide and pyrites begin to decompose, while pure tin and lead have melted. At 500°C sulfide ores (in air) begin to roast, while coldworked copper and bronze have fully recrystallized and become soft. At 600°C clay pottery develops moderate hardness and some types begin to develop a vitreous finish. Some glazes are molten at this temperature, but more heat is required for metallurgical slags to run properly. At this or even at a lower temperature, it is relatively easy to reduce copper, lead, and iron from pure oxide minerals by contact with charcoal; but the reduction of an ore is not metallurgically practical unless the metal can be separated from the rocky impurities. This requires that the metal be melted, and it is usually also necessary for the ore to reach a temperature high enough to fuse the earthy matter contained in it. The common early slags for all metals were composed of iron silicates or calcium-iron silicates melting at about 1200°C. Copper, lead, and tin conveniently melt below the temperature of fusion of the slags; iron does not melt when the slag does (its melting point when pure is 1539°C), but even at a dull red heat it is reduced to a metallic sponge which can be easily consolidated by hammering to weld the particles together when the slag melts and can be expelled.

Early pyrotechny was in fact a case of unity in diversity (39); the metallurgist and potter utilized each other's products and wastes. Metals or their oxides form the chief coloring matters in pottery and glazes, and the metallurgist had much to learn from the potter about the character and reducibility of oxides. At the same time the potter needed only to look at the slags of a reducing hearth to discern glaze and glass, which incorporate silica and alumina, the wastes of metallurgy but the materials of ceramics. When the smith needed a crucible he got it from the potter.

There unquestionably were hiatuses in communication, produced by the tendency for trades to stick to their own sections of the bazaar and to have their

own secrets. Glazes seem to have originated more in raw ores or the wood ashes of the potter's kiln than in slags; and there was an unnecessary thousandyear lag before the potter learned to make glass. But supremely important common knowledge did emerge. The craftsmen learned (i) that oxidizing and reducing atmospheres are almost as important as temperature-witness their effects on the color of a pot or a glaze, or in the fire refining of copper; (ii) that slagging of glass-like impurities is the crucial condition for successful smelting of most ores; (iii) that unrestrained temperature might be disadvantageous, whether in roasting ores, melting a metal, or baking a pot; (iv) that admixtures of other elements lower melting points, whether of glass or met-



Metals as art. (a) Bronze mountain deer. Length, 5.3 cm. (b) Bronze humped cow mounted on four wheels. Length, 11.5 cm. (c) Bronze oxen with yoke and plow. Overall length about 35 cm. These items are from the recent Marlik excavations in Iran (about 1200-1000 B.C.)

4 DECEMBER 1964

als; and (v) that heat has highly variable chemical accompaniments.

With an understanding of these connections between the pyrotechnic arts. one can better appreciate the moments of historic breakthrough suggested in Table 2, as pottery, metallurgy, and glazing were developed. There is a relationship between evidence that the potter of the 5th millennium possessed the closed kiln and evidence that the smith could melt copper. It seems more than fortuitous that Ubaid pottery, baked at high temperatures under reducing conditions, appears around 3500 B.C., concurrent with the first extensive smelting of metals. Nor is one surprised that the art of glass-making originates in Egypt simultaneously with the invigoration of the metallurgical industry under Mesopotamian influence.

#### **First Scientific Consequences**

Though man thus mastered the complex skills of practical pyrotechny, its scientific lessons eluded him for many millennia. It may be said that artisans discovered the metals and the metallurgical processes in a practical way, but did not identify them at all scientifically (40). The pseudoscience of alchemy was a natural consequence of the first dazzling successes in transmuting base matter to metals. Even nonspeculative workers continued to regard tin, antimony, and arsenic as having some connection with lead and retained such Latin terms as plumbum candidum (tin) and *plumbum cinereum* (antimony) until the time of Agricola. Steel was not understood at all. The word kassiteros originally meant ores containing no tin, as well as ores containing tin. As R. J. Forbes has shown (1) the confusion penetrated as far as Africa.

Indo-European etymology shows the effect of the first encounters with Asian metallurgy, though the Indo-Europeans made their first appearance on the plateau very late, possibly early in the 2nd millennium. The modern Persian ahaen, iron, has several possible roots (41). The word may be derived from the root ai, from Sanskrit ayas, which yields the Latin aes (copper) and, some scholars believe, the German Eisen and the English iron. Or it may be related to a group of words with as stems: asan, the Sanskrit for stone, and asman, a parallel word for sky. The etymology may indeed be connected with the

Homeric notion that the sky is a metallic bowl.

Not until the 18th century A.D. did Western man finally begin to slough off his misconceptions and appreciate the complex chemistry of metallurgy. Even at that time many reputable scholars continued to believe that certain irons have a "coppery" quality.

Our enlightenment about the structural and physical properties of metals has been even slower. Serious metallography is only a century old, and x-ray diffraction, which elucidated the atomic structure of metals, has just passed its fiftieth birthday.

# The Spread of Metallurgy

One must doubt that the tangled web of discovery, comprehending the art of reducing oxide and then sulfide ores, the recognition of silver, lead, iron, tin, and possibly arsenic and antimony as distinctive new metallic substances, and the technique of alloying tin with bronze, could have been spun twice in human history. But the matter is so exquisitely involved as to bear much further scrutiny.

Pyrometallurgy came into being progressively at several sites in a thousandmile zone across Anatolia, northern Iraq, and Iran. The arts of hammering and annealing native metals had already been widely known for a millennium or more in the southwestern Asian plateau and were likewise to become the first metallurgical arts in nearly every major metalworking culture from Hungary to Britain in the West and from eastern Asia to the Americas in the East. Whatever the case for the independent discovery of these arts on each continent, the discovery of full smelting with fire was almost surely transmitted from one culture to another. This is revealed by the wide prevalence (at least in Europe) of a single pattern of development: The earliest coppers are very pure; in later coppers impurities are found, an almost certain indication of the advent of smelting; finally tin is given priority as the alloying element. In China only the first stage of the sequence is omitted. This parallelism in the development of pyrometallurgy bespeaks not a series of separate births but remarkably sensitive technical communication out of southwestern Asia under most primitive circumstances.

Other evidence of such early techni-

cal communication is presented both by the movement of agricultural crops between civilizations of the Eastern and Western Hemispheres and by trade in minerals ranging from obsidian to tin. Trade may, in the case of scarce tin, have leaped westward to Spain and Britain and eastward to India and China, introducing men to mining before they knew metals with any precision. Subsequently, adaptation and selection gradually occurred as inevitable concomitants of this diffusion. Egyptian glass may have been an indirectthough sensational-result of the Mesopotamian trade in metals. The Chinese became pre-eminent in casting both bronze and iron, a fact which is often cited as proof of the independent discovery of both metals but which really indicates experimentation with known processes.

The great waves in which the invention of pyrometallurgy spread had diminished to small ripples by the time they reached the New World. Bronze technology gradually petered out as it spread northward from a center of implantation (perhaps in Peru), and terrestrial iron was utterly rejected by the pyrotechnicians of the New World. In China iron was known almost exclusively in its cast form, but in the Western Hemisphere it was not adapted at all. In Africa, by contrast, the age of bronze was skipped entirely, and iron in time became the dominant metal.

Clearly, one must draw a careful distinction between man's awareness of a metal and his ability to exploit it industrially. Copper so dominates the early history of metallurgy that the casual student may not be aware that its subtle, gradual, and logical progression from its native state to cast, industrially reduced, and alloyed forms was an exception rather than the rule. Lead stands at the other pole as a soft, ugly, pernicious metal which first manifested itself in the tribesman's hearth with practically no antecedents. Men eventually found uses for it, largely in yielding silver, but these uses were not important enough to dominate a stage of civilization. Even the archeologists have forgotten to look for it.

But iron nearly suffered a similar fate, as the history of the Western Hemisphere shows. The world owes a great debt to those quiet technicians— Hittite, perhaps—who sufficiently overcame their predisposition in favor of casting metals to resolve the problem of over-absorption of carbon by iron in the fluid state and to put iron to general use as a wrought metal. But Chinese bronze casters who immediately saw the virtues of cast iron are to be acknowledged equally.

In sum, the metal "ages" provide useful categories for the broad sweep of history; but they tell nothing about the true origins of metallurgy.

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