Chapter 2
The Historical Development of Metallurgy

The utilization of metals and the development of metallurgy has been an important societal progress for mankind: the introduction of metallurgy was fundamental to the evolution of complex and almost hierarchical society systems. The evaluation of the real impact of metallurgy on ancient societies has been quite an intellectual challenge to archaeologists and historians without substantial data of metallurgical processes and innovations, properties of metals and alloys, information about mines and the distribution of raw material.

Forbes [11], a historian of technology, has introduced his book *Metallurgy in Antiquity* with the sentence "…facts which can be gathered from archaeological, historical, philological and technical documents and books are not sufficient to form a continuous story". Nonetheless, he wrote the book "with fancy as the woof of the story".

Indeed the research into ancient metallurgy is not a new discipline, but systematic interdisciplinary investigations during the last half-century generated a wide range of perspectives on that field called archaeometallurgy (see Craddock [8]; Roberts and Thornton [37]; Timberlake [46]). Archaeometallurgical research is not only based on the chemical, structural and technological characterization of metals themselves but includes the investigation of all pyrotechnical debris such as slag and technical ceramics: crucibles, moulds or furnace linings to reconstruct ancient metallurgical processes (see, e.g. Craddock [6]; Roberts and Thornton [37]). The characterization of ore deposits and their minerals is also an important task to correlate artefacts with their provenance and to reveal trade and social relations of the past. Mining archaeology has achieved sustained success in rediscovering and dating ancient mines, in the reconstruction of the mining technology, the quantitative output and the interaction with the surroundings (e.g. Domergue [10]; O'Brien [27]; Tripcevich and Vaughn [47]). Finally, the continuous (re-)evaluation of written sources and of historic graphic material in the view of new research is still an important task. There have always been some key issues, such as the beginning of metallurgy within certain regions, the earliest use of metals and alloys or the transition from one technology to another. Nevertheless, the march of historical usage often

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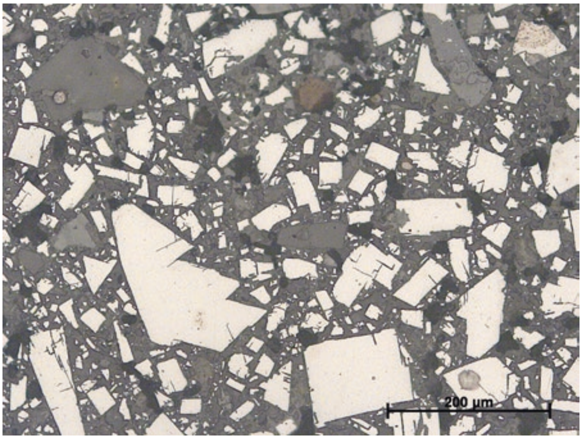

Fig. 2.1 Polished section of a so-called lead bead from Neolithic Tell Halula (Syria) viewed in bright field showing high reflecting cubic galena (PbS) crystals with cleavage within lead carbonate

exposes the lack of knowledge concerning properties of individual alloys and their deliberate use or production in different regions during specific periods.

The use of metals goes back some 7000–8000 years. In the Old World, we move from a premetallic phase where minerals were used for gemstones to a Chalcolithic copper phase. Copper was the first metal used by humans and not lead, as far as we can see today. Some incorrectly labelled lead minerals from Çatalhöyük [34, 43] and some other places (Fig. 2.1) have introduced much confusion into this discussion, which is not finally closed (see Craddock [6, p. 125]). Indeed Moorey ([25, p. 295]) maintains that lead was one of the first metals to be used by man, although he used to say that it was *the* first!

It is usually an easy task to distinguish between minerals and metals by metallography, whereas field analytics, for example, with portable X-ray fluorescence devices (p-XRF), which are very popular today and generally helpful, can be quite misleading, because it is not possible to detect many light elements which are essential components of mineral compounds. Indeed, with increasing sophistication and the use of portable vacuum or flushing techniques, even sodium and magnesium can now be seen, even if not easily quantified.

Arsenical copper was the first alloy deliberately used by man, but bronze has most probably been the first alloy that has been deliberately produced. The archaeological period known as the Iron Age is different in its technical stages, in which we are still enmeshed, although steel itself is giving way to new composites, polymers and high-performance and nano-engineered materials. It is an interesting fact that iron and steel production by direct smelting, the so-called bloomery process, has been a valued technology in most parts of the world, which hasn't changed much for more than 2000 years. Whereas in Europe the introduction period of the indirect process, the blast furnace, in medieval times had quite a few teething troubles until the seminal improvements in the nineteenth century. In ancient China these concerns were obviated by the introduction of white and grey cast iron long before the west began to make use of it.

The concept of continual progression and societal development is firmly entrenched in this series of transitions from the hammering of small pieces of native copper to the production of alloyed steels and beyond. It is a linear view that disguises a number of truths about metallurgical development in different continents and different societies, connecting style, systems of belief and technological choices (see Lechtman [21]).

In the local development of different metallurgical traditions, independent inventions, skills introduced by diffusion and local experimentation are the reality of the complex mix of events, which make up the metallurgical past. There is most probably not just one story but many intertwined stories with different origins at different times. This, surely, should be an obvious facet of human history, but some archaeologists, in applying broad concepts to the development of metallurgy, have tried to simplify this complexity, arguing against the "diffusionist" hypothesis of an earlier generation of archaeologists to expand on the theme of "independent invention", which does not explain the complex tapestry of the metallurgical past.

Childe [4] puts forward the idea that metallurgy originated in the Near East and was adopted in Eurasia by diffusion, whereas Renfrew [35] argued for the independent invention of metallurgy at different places such as southern Iberia in the fourth millennium BC. This is a view which has been thought to lack evidence but which is also still in dispute because of more recent scientific evidence from Europe. The discussion about diffusion versus independent invention is reviewed by several authors in *Archaeometallurgy in Global Perspectives* [37]. Clearly, the *ex oriente lux* theory is an oversimplification of the complex societal events involved in the inception of metallurgy. The transubstantiation of minerals to copper metal and the early use of native copper have long been a source of fascination in attempts to unravel this mystery: Childe [5] talked of "homotaxis", or modes of development, with this primary period of copper use corresponding to "mode zero" of our development over time, while Daniel [9] talked of an "eochalcic episode". These neologisms, which now seem rather artificial, have faded into obscurity. More colloquially, and meaningfully, a whole age of humanity's past has been designated the Bronze Age, signifying the great importance of copper and copper alloys in a crucial stage of our development from hunting-and-gathering communities to settled villages and the rise of agriculture.

There are complexities regarding the early use of metals in the Old World, problems regarding the validity of the independent invention model, the transmission of metal objects, metallurgical practices and the role of metallic artefacts [44]. New discoveries are continually shifting the picture that we have of the earliest use of copper (Chap. 5). The earliest evidence for native copper and copper oxide use for beads, hooks and needles in Europe is now thought to date from the mid-sixth millennium BC, from sites such as Divostin and Rudna Glava in Serbia [1]. Rudna Glava itself is most probably an enigmatic example of error or of misinterpretation in the research into ancient metallurgy. Rudna Glava is without doubt one of the oldest copper mines in Europe, with dates that show that the mine was in use from the late sixth millennium BC [1, 27, pp. 40–7], and it has been assumed that this mine has been the major source for Eneolithic copper of the Balkans (e.g. Jovanović

[15]). Indeed copper oxide minerals such as malachite were won, but as of 2019, perhaps only one single Neolithic copper artefact from the Balkans has been shown by lead isotopic analysis and chemical analysis to derive from the ore minerals of Rudna Glava [12, 29, 30, 33, 38]. It might be possible that further analyses could indicate the use of ore minerals from Rudna Glava for early copper smelting, as Gale and his co-workers [12, p. 169] have already suggested, but at the moment it seems more likely to be a misinterpretation of archaeological evidence for copper mineral winning.

However, the contemporary settlement of Belovode, also in Serbia, provides the earliest documented proof of pyrometallurgical copper extraction in Europe [33], which is at a comparable date to sites in Anatolia (Chap. 5). In other areas of Europe and the central Mediterranean, there is increasing evidence of copper mining and smelting from the late fifth millennium BC onwards [2, 27].

It is obvious from many lines of research that smelting of the copper ores would have been initially small scale, operating at relatively low temperatures, carried out under poorly reducing conditions using oxide or roasted sulphide ores in small stone or clay structures or crucibles without intentional flux additions, creating relatively little slag [2, 7]. Smelting of this kind usually creates prills of copper which have to be further refined (Fig. 2.2), often by remelting in a crucible. Some of these prills, which are usually totally transformed to oxide compounds, are sometimes found in settlements, and there is again a serious confusion in terms of natural copper mineral compounds or corroded copper.

Our ability to provenance early copper sources is limited, because many old mines have been eradicated by modern mining or not yet identified. The general approaches and their benefits or failures to link ore to metal have been controversial

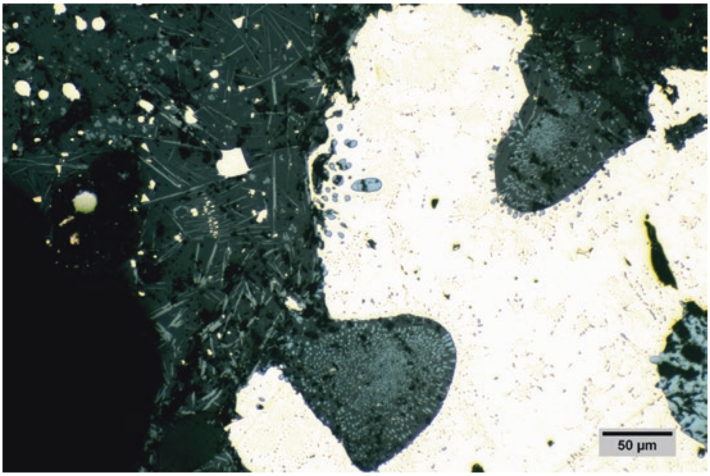

Fig. 2.2 Experimental smelt of copper from high-quality malachite, smelted using a simple clay mini-furnace and charcoal with hand-held bellows used for heating. The result was prills of copper entrapped in a tenacious slag. The slag incorporates some fayalite in a glassy matrix

and have been discussed many times elsewhere (for references see Pernicka [28]; Pollard and Bray [32]), but thorough case studies, such as those from the Balkans, the Mediterranean and other regions have clearly demonstrated the successful application of scientific methods to such questions.

A good example of trial and error, combined with basic research in archaeometallurgy, is the question of the origin of Eneolithic material in the British Isles and continental Western Europe termed Bell Beaker metal. More recently Roberts [36, p. 433] has referred to the general problems of source definition of the Beaker metal, which remains a problem. A case study focuses on the British Isles: Junghans and his co-workers [13, pp. 128–138], part of the so-called SAM group (see Pernicka [28]), have selected a group of arsenical coppers, which is prevalent on Ireland and in some regions of Central Europe during the third millennium BC. Due to the lack of knowledge concerning contemporary copper mines on Ireland and maybe due to the continental influences on the Beaker culture, they suggested an import of metal from Central Europe. Further analysis could show that some of the material also reached Britain, but now Ireland has been proposed to be the main source, which has also been questioned (see Northover et al. [26]). With the discovery of the arsenic-bearing copper mines on Ross Island, it has become clear that it was one of the major sources for copper for the British Isles in the late Chalcolithic and the Early Bronze Age [26, 27]. The SAM group has misinterpreted their data, because of the lacking knowledge, but as a result of their work, they had predicted the Ross Island material through their basic research, several decades before the discovery of the mine itself.

By the second millennium BC, when tin-bronze became common across much of the Old World, there is a significant rise of copper mining and metallurgy, even on quite different scales. Small-scale mining sites such as Copa Hill in Wales, dated 2100–1600 BC, show that advances in this kind of technological process had reached quite far-flung locations with many logistical problems [27, 45]. Most of these early mines were run by open-cast mining to extract weathered secondary copper minerals, but some mines show a complex series of vast underground tunneling [3, 6, pp. 23–92, 10, 27]. The shafting of Kargaly in the southern Ural has reached around 40 metres depths, but the tunnel system embraces a territory of nearly 500 km^2 [3]. The Mitterberg area in the Tyrol is one of the largest Bronze Age mining fields in Europe and shows mining galleries with nearly 200 m depth [31]. With increasing demand and more underground mining, a drastic change in ore choice from oxidized secondary to sulphidic minerals occurred, which involved more complex technologies for ore processing and smelting.

Most Continental European Early Bronze Age artefacts clearly show fahlore impurity patterns (see, e.g. Krause [20]), whereas Middle Bronze Age metal generally has lower concentrations of impurity elements like antimony, silver and bismuth [31]. The Mitterberg mining area lodes are dominated by some Ni-As minerals and chalcopyrite, which is connected with a three-step matte smelting process to remove sulphur and iron by roasting and slagging, before the copper can be reduced. Iron-rich slag containing matte prills is the common metallurgical relicts of this eastern alpine process [19], but similar debris can be found from many sites all over the Old World (e.g. Hauptmann [14]; Kassianidou [16]; Zwicker et al. [52]), albeit they are not necessarily evidence for a deliberate matte smelting.

Early iron smelting may originate from the smelting of iron-bearing copper ores or from the addition of iron-containing fluxing materials under highly reducing conditions in the smelt. However this remains uncertain, although it is a convenient model to explain the advent of iron, with quite a lot of indications from the archaeological material as to the likelihood of this (Chaps. 4 and 5).

The transition from bronze to iron in the different parts of the world is actually an interesting topic for many economic and technological reasons (Chap. 5). Nonetheless, innovations do not follow a simple logic, and the triumphal procession of iron has undergone many vicissitudes in different parts of the world, from the appearance of the first iron implements to full-fledged iron using societies (see Wertime and Muhly [50]). Cast iron technology first emerged in China from the ninth century BC and has been transferred to other Asian countries many centuries later, but it took around 2000 years until European bloomery smelters changed to this technology, while African iron smelters never did.

Each continent has its own especial creations or metallurgical developments depending on the culture and the geography, the ores available in the region concerned and how these ores are utilised or processed. Thus, early metallurgical development in Africa encompassed around 11,000 km of distance from Egypt to the Cape and nearly five millennia [17], with the principal metal produced being iron. The advent of copper and iron metallurgy on the African continent is an interesting topic, and especially in sub-Saharan Africa, the beginning of the use of iron is one of the most debated features of the metallurgy, as there is no bronze metallurgy preceding this important development, essential for so much of African agricultural development (see Van der Merwe [49]). Some practices extend through time remarkably, for example, isolated human finger bones were buried beneath prehistoric furnace bases in the Lowveld of eastern South Africa to ensure a successful smelt, and the same practice is recorded by the early twentieth century ethnographers in the same region.

For various reasons, not much is currently known of early Egyptian iron smelting compared with sub-Saharan Africa, perhaps because excavations are more focused in these areas on iron metallurgy and more readily accessed. Copper objects first appeared in the Maadi culture near the Nile delta between 4000 and 3200 BC. Copper and gold artefacts first appeared in lower Nubia and are dated from 3600 to 3300 BC, the increased use of metals being a long slow process. Surprisingly, there was no tin or bronze, no silver, no gold and no lead in southern Africa before the beginning of trade with the Islamic world [17]. The lack of these developments may have a bearing on the discussion concerning the independent development of metallurgy in Africa, which could well prove to be an indigenous technology as was the entire metallurgical technology of South America. As far as ferrous metals are concerned, their origin in Africa has been something of a mystery. There are several very old radiocarbon dates in the literature, and recently Zangato and Holl [51] have suggested that iron production activities have taken place in Central Africa as early as

3000–2500 BC, which would be the earliest iron working in the world (see comments in the same issue of the journal) which is further discussed by Killick [18].

In the New World, metallurgical development had not yet reached the stage of the extraction of iron by the fifteenth century AD, but given the parallels between human inventiveness in the Old and the New, there is no reason to think that iron metallurgy would not have come sometime after 1500 AD, when the continent was invaded by the Europeans and the native cultures destroyed.

In broader terms there is "independent invention" of metallurgy in the New World, which shows that metallurgy can be developed without contact with cultures which already have discovered metallurgical processes. The necessary materials required, ceramics, clay, charcoal, stone, wood and fire, are universally available. These materials needed to come first, especially ceramics and the ability to shape and fire large ceramic structures or make refractory crucibles in which metals can be manipulated or smelted.

Extractive metallurgy in the New World began in the Southern Andes, where the mineral resources are rich in copper, tin, lead and silver (see Tripcevich and Vaughn [47]). Extractive metallurgy developed strongly in this region and "diffused" up towards the North, with the dates for metallurgical activity in Costa Rica, being later than those in Colombia, which are later than those in Ecuador and Peru. The separation between different cultures in the New World resulted in the smelting of sulphide ores being prevalent in Peru but absent in Colombia [22, 23, 39, 41]. The pre-eminent metallurgy of Colombia, Panama and Costa Rica is that of gold-copper alloys often with depletion gilding and a variety of different coloured gold alloys, especially during the period from the early centuries AD onwards (Chap. 6). Silver was not extracted in this region and occurs in the gold alloys as a result of its inclusion in the native gold but was extracted in Peru and Chile, for example, where extensive use was made of silver.

Copper may have been smelted from oxide ores, but not from sulphide ores in the Colombian area, while in Peru, complex metallurgical extraction processes had been developed for the cupellation and scorification of silver, the alloying of silver-copper alloys, the fabrication of silver-gold-copper alloys with extracted silver and the extensive use of copper-arsenic alloys. One of the remarkable alloys made for decorative purposes is an extensive range of silver-copper alloys, which extends into Ecuador, but only crosses the Colombian border as far as the extreme south, in the Nariño region. These alloys are often surface depleted in copper, resulting in heavily debased silver-copper alloys which appear to be made of silver. This decorative use of metals is an important one: metals were not primarily instruments of war, in the same way that they were in the Old World. Bronze Age peoples made extensive of tin bronze for arrowheads, swords, daggers and knives. Copper alloy mace-heads are the most well-known South American war-like metallic implement, which we associate with the Inca Empire, but in earlier cultures in South America, the extensive use of metals for this purpose was practically unknown.

Many unique metallurgical achievements took place in South America. Perhaps the most impressive is the development of electrochemical replacement plating in Peru to deposit very thin coatings of gold or silver onto copper [24]. The other is the use of platinum plating or cladding in the La Tolita-Tumaco culture of Ecuador, which before the birth of Christ had already made use of platinum to produce platinum-gold composite artefacts, at a time when platinum was totally unknown in Europe [39, 40]. Native platinum from South America first entered the European market during the eighteenth century as "white gold" and attracted considerable interest because of its difficulties in working or casting to shape [48, p. 152]. From the eighteenth to the nineteenth centuries, the number of metals known increased significantly, but many others such as arsenic, antimony, nickel or zinc have not been known in metallic form before modern times or only sporadically in their native forms, but some of their alloys enjoyed great popularity from prehistory (Chap. 5). Indeed metallic zinc has been used in Asia from the thirteenth century AD onwards, but it has not been produced in Europe before the eighteenth century; nevertheless brass has been the most popular copper alloy from the first century BC until today (Chap. 5). Actually, we do not know if brass making has been developed independently in the Old World or has been transferred from Asia or vice versa. Certainly, we can say that the indigenous people in the New World did not undertake the production of brass and therefore have not used it before the first Europeans entered the continent.

We have seen remarkable progress in understanding ancient metallurgy, but there are many gaps in terms of knowledge and comprehension about individual metallurgical practices and developments. Metallography has been an essential part of the unravelling of the story of these metallurgical developments. The identity of an excavated artefact, even if heavily corroded, can usually be determined by metallographic examination. The mounted section can then be used for further, detailed studies to determine precise compositional data. Determining whether an artefact has been cast or worked to shape is most important for the history of metallurgy, and this kind of information has been slowly assembled through thousands of studies carried out over the past 100 years. During that time, the fine achievements of pattern-welded swords and damascened iron were unravelled; the Chinese story of grey and white cast iron and the history of the utilization of copper, arsenical bronze and tin bronze elaborated; the subtleties of the extraction of silver and the use of silver alloys defined; and the use of tin, gold, silver, bronze and platinum as plating or cladding materials investigated, and numerous site reports have been issued, containing metallographic images and reports which are essential for our understanding of the archaeological past. The chapters which follow provide a brief overview of some of the important developments in the field of metallography of ancient metals.

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