Coins, artefacts, and isotopes – archaeometallurgy and Archaeometry

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Abstract

Archaeometallurgy is one of the earliest manifestations of archaeometric research, using science-based approaches to address cultural-historical questions. This review first outlines the extent of the field, defining in some detail the main branches of archaeometallurgy, and their specific methodological approaches. It then looks at some of the early publications pioneering archaeometallurgical research, to set the scene for the publication pattern of archaeometallurgy in general, and the role Archaeometry played in publishing archaeometallurgical research. The analysis of archaeometallurgy-themed publications in Archaeometry, their change over time, and their relationship to the total range of work done in the field, indicates that there is a rather narrowly defined and specific type of archaeometallurgy which gets published in Archaeometry, initially with a strong focus on coin and object analysis, often combined with method developments. The more recent developments in isotopebased studies in archaeometallurgy find only a limited representation in the journal, despite the leading role which the Isotrace Laboratory played in this discipline, for some considerable length of time. More recently, this Archaeometry-specific 'flavour' of archaeometallurgy seems to weaken, with an increase of papers on iron and on primary production in general, subjects still much under-represented.

Keywords: Archaeometallurgy, coins, isotopes, publication pattern

Introduction

Archaeometry as an academic field can trace its roots back to several diverse academic 'families'. Created through the marriage of physical and historical sciences fifty years ago, archaeometry combines specialised applications of science-based approaches to archaeological and historical questions, with sub-disciplines such as geophysical prospection and remote sensing, absolute dating, ceramic studies, geoarchaeology, archaeobotany and archaeozoology, and archaeometallurgy. The establishment of the journal *Archaeometry* in 1958 served as a milestone in the formalisation of archaeometry as a mature scientific discipline; but how successful has it been in catering for the needs of the various sub-disciplines?

As is the case with many other sub-disciplines of archaeometry, archaeometallurgy has evolved into a subfield in its own right. A number of journals now specialise in archaeometallurgy, and numerous conferences exclusively devoted to metals in

antiquity have been organized since the 1980s. However, *Archaeometry* was the first journal at the interface of natural and historical sciences, and has long remained its backbone. But by and large, archaeometallurgy only played a minor role in the topics covered. This, clearly, has something to do with the mother institute behind the journal, the Research Laboratory for Archaeology and the History of Art (RLAHA), and its offspring, the Isotrace lab. We leave it to others to present and discuss the formal and historical aspects of the relationship between these three. Here, we focus on a view from the outside, on the impact the journal has had over the last half a century on the study of ancient metallurgy. To do so, we need to sketch out the size, shape and content of the 'field' archaeometallurgy before assessing its major research outlets, and the role that *Archaeometry* played in this.

What is Archaeometallurgy?

Metal objects play a significant role in most post-Neolithic societies, as reflected in the denominations for major archaeological periods (Copper Age, Bronze Age, Iron Age). The sequentiality of these units reflects the perceived stepped introduction of major metals and alloys, spanning from the earliest use of a few native metals (mainly copper) probably some 10,000 years ago, and continuing to this day with the development of ever more sophisticated alloys based on the more than 70 different metals in the periodic table of elements. Broadly speaking, archaeometallurgy deals with all aspects of metal production, distribution, and usage in the history of mankind (Fig. 1). Archaeometallurgists often concentrate on periods before c. AD 1500, when only seven metals were known and a number of their alloys, i.e. gold, copper, lead, silver, tin, iron, and mercury, and the alloys of copper (copper–arsenic, copper–tin, copper-tin-lead, copper-zinc), silver (silver-copper, silver-gold), pewter (tin-lead) and iron (iron-carbon, iron-phosphorus), although there are good reasons to extend archaeometallurgy to much more recent periods (e.g. Goodway and Odell 1988; Gilmour and Northover 2003; Rehren 2006; Bourgarit and Plateau 2007). Thus, it is not primarily the age of the material studied which defines archaeometallurgy, but the application of specific methods to address cultural-historical questions. Reliance on scientific methods is often dictated by the 'ahistorical' nature of the crafts well into the recent past, resulting in at best patchy contemporary textual documentation being available.

The first use of metals some 10,000 years ago was from natural occurrences as native metals which did not require elaborate mining and smelting. This early metallurgy is limited to specific geological areas and typical of the earliest use of gold, silver, copper, iron, and mercury. The use of these native metals initially followed earlier, rather mechanical, approaches to lithic materials. However, supply of metals increased dramatically with the inception and spread of mining and extractive metallurgy, the origin of which is not yet clear but seems to have risen in the late sixth millennium BC; at the same time, genuinely metallurgical production and manufacturing techniques were developed, considerably expanding the use and versatility of metals. The emergence of alloys, both natural and intentional, further widened the range and appeal of metals available. In spite of these innovations and the subsequent, almost global, spread of metallurgy, the geological limitation of metal production to areas rich in specific ores remained. This necessitated or stimulated developments in other fields such as economics, politics, warfare, or trade, to match the spread in knowledge with a similar spread of the material. Many civilizations

flourished in areas devoid of metal ores, such as the large river valley cultures of Egypt, Mesopotamia, and the Indian subcontinent; other areas were rich in one metal but not another. Thus, mechanisms of trade and exchange, and methods for the recycling of or substituting for metals were of considerable importance from an early period onwards, and their study is firmly within the realm of archaeometallurgy.

A major division separates the primary production of metal from the manufacturing of artefacts, namely the nature of the skills required for each. Primary production, i.e. mining, beneficiation and smelting, requires a keen eye for specific minerals, their relevant properties such as colour, hardness, smell, mechanical behaviour under stress, chemical behaviour at high temperatures etc., and knowledge of the necessary associated materials such as fuel, technical ceramics, and fluxes. The archaeological evidence for this type of activity is mostly waste material such as slag, furnace and crucible fragments. Manufacturing, on the other hand, requires a fine understanding of the behaviour of metals and alloys, over a range of temperatures from cold to fully liquid, combined with the artistic skill required to make both the functional and beautiful objects desired by patrons. Similarly, archaeometallurgy separates into a number of parallel main strands, based on the nature of the materials available for study (waste vs. artefact), the skills involved (smelting vs. manufacturing), and the archaeological context (workshop sites vs. consumer sites). Of course, the division into primary production of metals and manufacture of artefacts is not absolute, and considerable overlap exists between the two strands; indeed, this division took some time to develop, and much of the earliest evidence suggests that the first metallurgists were covering the entire metallurgical *chaine operatoire*, from ore prospection to artefact production. This division does hold true, however, for the bulk of archaeometallurgy, and is mirrored in different analytical approaches, reflecting the different nature of the materials involved and questions asked. We will use this division also to structure the following review.

It may be added that especially the primary production has a strong overlap with a third strand, namely mining archaeology, the investigation of ancient mines with archaeological methods of documentation, excavation and typology. However, since papers of this subject are not represented in *Archaeometry*, we will not deal further with it. Similarly, we will not attempt to cover the other end of the archaeometallurgy cycle (Fig. 1), the corrosion and conservation of artefacts during and after burial, aspects which are also not typically covered by publications in *Archaeometry*.

Primary production of metals

The geological link between the often remote ore deposit and the main production sites, with their evidence for smelting operations to extract the metal from the ore, offers a unique window into the activities carried out, away from the typically urban consumption areas. Almost universally, the waste materials remained in the immediate vicinity of the site of production, providing reliable evidence of the activities and technologies employed here. Metallurgical activity leaves three main types of evidence: metal as raw lumps and spills, semi-finished products and objects for repair or recycling; associated products such as discarded ore, slag, matte and speiss; and remains of tools and installations such as crucibles, hammers, tongs, furnaces or hearths. Of these, slags are typically the best preserved, most abundant and most informative. However, they are also least accessible by traditional archaeological methods such as typology or stylistic analysis, but require scientific

analysis and expert interpretation to reveal the information they contain. Even the most fundamental of identifications are not always possible using field methods and visual inspection. Metallurgical slag can be confused with geological material, or artificial materials from processes other than metallurgy. The differentiation between primary production or smelting on the one hand, and secondary production or reworking on the other hand, is often indicated by the wider archaeological context, but this cannot be taken for granted. It can be difficult to distinguish in the field between ferrous and non-ferrous metallurgy, or between iron smelting and smithing. However, studying the waste material can yield very specific information about metallurgical processes and ore types, production technologies, and scale of production. Identifying and understanding these aspects of production is at the core of the archaeometallurgical analysis of slag.

The production and working of metal is controlled by two main factors: technical constraints and cultural traditions. While there are certain fixed physico-chemical conditions to be met for specific metallurgical operations such as smelting, alloying, refining, casting, and recycling, there are many different configurations which may meet these conditions. The composition and quantity of the resulting materials, primarily metal and slag, reflect both factors. It is by identifying the fixed physico-chemical constraints that the culturally determined configurational factors can be revealed, producing archaeologically relevant information (Rehren et al. 2007, and literature therein).

The first issue addressed by slag analysis is the identification of the type of metallurgical process which created it, and the metal and ore type smelted or worked at a given site. Ore deposits comprise two complementary materials: the rich mineral and the gangue or host rock. Ore beneficiation mechanically separates the rich mineral from the gangue. By smelting the metal is then extracted from the rich mineral through a series of chemical reactions while transforming remaining gangue into slag. Depending on circumstances, other waste or intermediate products form, such as matte (metal sulphides) and speiss (transition metals combined with elements of the fifth main group of the Periodic Table of the Elements, mainly arsenic and antimony). The type of ore, such as oxidic, sulphidic, or complex, is at best broadly reflected in the composition of the smelted metal. The slag, however, contains all the gangue components as well as components of the rich mineral, modulated by the smelting conditions. In effect, the waste gives a much more complete representation of both, the ore body and smelting conditions. This picture is complicated through the addition, conscious or not, of further material to the slag, such as fluxes, eroded furnace wall material, and fuel ash (Serneels & Crew 1997; Kronz 1998; Crew 2000; Veldhuijzen & Rehren 2007).

This leads to the identification of the nature of the operation. Metallurgical processes require elevated temperatures, typically in the range of 800 °C to 1400 °C, and a wide spectrum of redox conditions, spanning from highly oxidising to strongly reducing. Each metallurgical process has its own characteristic combination of temperature and redox condition. Neither can be determined directly, but both find their direct expression in the mineralogical make-up of the slag. Identifying these parameters is crucial for the basic identification of the technological process, as well as for identifying its particular configurational aspects, and relies heavily on mineralogical analysis (e.g. Bachmann 1982).

Finally, production remains are often well preserved and the best available indicator for the scale of operation of a given workshop or smelting site. Careful determination of total slag quantity and composition, in combination with an assumed or directly determined ore quality, can provide good estimates of metal production quantities by using mass balance calculations. Similar estimates can be made for workshop remains such as crucibles (Rehren & Papakhristu 2000) or smithing debris (Crew 1991; Serneels & Perret 2003); quantities can be determined either for a site overall, or on an average annual basis if the lifespan of the site or workshop is known. Such quantification is crucial for discussions of subsistence or surplus production, craft specialisation, and trade relationships.

Metal working and distribution

The use of metal objects can be seen as falling in one of three broad categories: decorative (jewellery, inlays and other accessories), military (arms and armour), and utilitarian (coinage, tools, general implements). These exploit the different metal properties perceptible in antiquity, such as colour, sonority, density, malleability, hardness etc. One reason to analyze metal objects is to understand whether for a given object these properties have been either selectively exploited, or even modified to suit the purpose. This information acquaints the archaeometallurgist with the state of metallurgical knowledge, or relative priorities of these parameters, of the person or society producing the object. Another reason for analysis is to discuss the functionality of objects, for example, whether funerary or dedicatory objects were made for display only or for real use. Reconstructing the techniques used to work metals by studying the waste left behind is a more process-oriented field which focuses on the workshops and their activities.

Chemical and in particular isotopic analysis is the main avenue towards identifying the geological origin of a given object, which directly addresses issues of trade and movement of objects. This begins with the desire to classify objects by material types and to identify similarities and differences in composition in order to form groups, using parameters which are independent of and often complementary to traditional archaeological typologies and art historical criteria. Finally, there is the necessity to identify the most suitable conservation methods to restore or preserve metal objects, and conservation science has its own important role to play within archaeometallurgy. Thus, four main research fields prevail in the analysis of metal objects: identifying their original composition and current condition, classification by compositional groups, reconstructing metallurgical practice from shaping (casting, mechanical deformation, etc.) and joining (welding, brazing, etc.) to finishing (decoration techniques), and locating the geological origin of the metal.

Analysing archaeometallurgy

The metallurgical analysis in archaeology has to be broadly separated into two main strands, one aiming to study metal artefacts, and the other concerned with the production waste. Both can use the entire range of analytical methods available, however, several approaches have been particularly successful and are therefore more widely adopted than others.

Primary production

Analysis of slag and ore in archaeology draws almost exclusively from earth science methods, primarily geochemistry, ore petrology, and igneous petrology. Ideally, this involves a multi-element method such as X-ray fluorescence (XRF) or inductively coupled plasma excitation with optical emission spectrometry (ICP-OES) in combination with optical and electron microscopy for the study of the texture of the sample and an assessment of mineralogical parameters. Ideally, the research question and aim should govern the choice of analytical method(s). In reality, costs of analysis and ease of access to or availability of instruments and expertise often play a decisive role in selecting methods of analysis. For all quantitative methods it is imperative to monitor and report data quality (accuracy and precision) through publishing results for analysis of certified reference materials along with the unknown samples, in order to be able to compare data from different laboratories.

Metallurgical smelting slag often occurs in huge quantities, accumulated over long periods and measuring tons or even thousands of tons. Sampling methods developed for earth sciences are often appropriate for stratified profiles and reducing large sample volumes through homogenisation and quartering into aliquots. Curatorial constraints are often more important in the analysis of other waste materials, such as crucible fragments, which have a stronger developed object character and typically do not occur in such large quantities. Here, cross sections prepared for Reflected Light Microscopy (RLM) and Scanning Electron Microscopy (SEM) with attached Energy Dispersive Spectrometry (EDS) are more suitable than bulk chemical analysis. SEM-EDS has relatively high detection limits in the order of 0.1 wt% for most elements, and therefore provides only basic chemical information; however, it offers a high spatial resolution of what is analysed, ideal for complex and multi-phase materials such as crucibles with internal slag coatings and external vitrification layers. A balance between the curatorial desire to minimize the sampling impact and the analytical need for a representative sample is sometimes difficult to achieve, and may require the use of non-invasive and non-destructive methods such as surface-XRF or micro-XRF (Tite et al. 2002).

Artefact analysis

For artefact analysis it is often important to use methods which do not alter the physical integrity of the metal objects, such as neutron activation analysis (NAA), Xray fluorescence analysis (XRF), proton-induced X-ray emission or gamma emission (PIXE or PIGE), or X-ray fluorescence analysis with synchrotron radiation (SR-XRF). NAA is not strictly non-destructive as the object can only be returned after the decay of the artificially induced radioactivity, and only relatively small objects can be irradiated in a reactor. Since some nuclides have rather long half-lives, it is necessary to initially determine the composition of the object by other means before the high sensitivity of neutron activation analysis be sensibly employed. A typical case is unalloyed copper. From the matrix element only short-lived radionuclides are formed and after a decay period of a few days up to 20 elements can be determined at trace levels (Hancock et al. 1991, Kuleff and Pernicka 1995, Rapp et al. 2000). After a few weeks the radioactivity has usually fallen below detectable levels. It is often important to analyse a sample non-destructively so that other methods can be used to determine other parameters on the same sample. A typical example is the combined trace element and lead isotope analysis of copper and copper-based alloys. An integral part

of good laboratory practice is the documentation of the analytical procedure, and the storage of part of the analysed material for future reference. This may sometimes lower the sample mass available and consequently increase the detection limits for certain elements for methods which require a sample to be removed and dissolved, such as atomic absorption analysis (AAS) and excitation with an inductively coupled plasma either for optical emission spectrometry (ICP-OES) or mass spectrometry (ICP-MS). Often the optimum choice are methods which remove a minute amount of material through evaporation or ablation, such as laser ablation ICP-MS or secondary ion mass spectrometry (SIMS). However, these instruments have rather small sample chambers so that only small objects can be analyzed. Larger objects require sampling which is often easy to carry out, unless for gold objects. In these cases relatively elaborate techniques like PIXE or synchrotron XRF are required for analysis.

Isotope analysis

In the 1960s, a fundamentally new method was arising from advances in geochemistry, namely the analysis of lead isotope ratios for the investigation of the provenance of metals (Brill and Wampler 1965, Grögler et al. 1966). The first tentative studies began to flourish in the 1970s fuelled by a collaboration of W. Gentner and G.A. Wagner at the Max-Planck-Institut für Kernphysik in Heidelberg and N.H. Gale at the University of Oxford. This group systematically studied the provenance of ancient Greek silver coins using both trace element and lead isotope analysis, combined with extensive field work on lead-silver deposits in the Aegean. The approach encompassed not only analyses of metals but also geological and mining archaeological field work, as well as mineralogical studies of ores and metallurgical remains (Gale et al. 1980, Wagner and Weisgerber 1985, 1988). A similar holistic approach, albeit without lead isotope analysis, was followed in the studies on chalcolithic copper metallurgy by E.N. Chernykh (1978). The breakthrough of provenancing by isotope ratios came with the extension of the lead isotope analysis to copper and copper-based alloys (Gale and Stos-Gale 1982). By the combination of lead isotope ratios and trace element patterns it became possible, for the first time, to relate with high probability metal artefacts to specific ore deposits, something that had been aimed at for more than a hundred years. The work culminated with major syntheses for the metal from Cyprus by Stos-Gale et al. (1997), and for the southeast European Chalcolithic by Pernicka et al. (1997). The merit of lead isotope studies has not been uncontested; a controversial paper by Budd et al. (1993) sparked off a discussion both in the Journal of Mediterranean Archaeology and in Archaeometry, with a number of comments in issue 35 (1993). However, it is now widely accepted and applied to a range of other materials such as glass and pigments (Lilyquist and Brill 1993; Shortland 2006).

Recent developments tried to exploit the isotope ratios of other metals of archaeological interest for provenancing, such as tin (Begemann et al. 1999), copper (Klein et al. 2004) or osmium (Junk and Pernicka 2003); so far, success has been limited. More promising seems to be a combination of metallographical, chemical and isotope analysis of iron (Schwab et al. 2006, Degryse et al. 2007).

A major problem is still the dating of metal by physical methods. There is often enough carbon in ancient iron to be measured by accelerator mass spectrometry, and this has indeed been used for dating purposes (Scharf et al. 2004, Enami 2004). However, Craddock et al. (2002) pointed out that carbon in ancient iron can derive

from various sources including geological ones like limestone that decomposes to carbon dioxide in the furnace resulting in erroneously high ages. For base metals the radioactivity of ²¹⁰Pb has been employed for authentication work (Pernicka et al. 2008). This method was originally introduced to archaeometry by Keisch (1967) for the authentication of lead white pigment in paintings. The only authentication method for gold based on the U,Th-⁴He dating method is just arising (Eugster et al. 2008).

It is interesting to see how for more than two decades much of this development was driven also by N. Gale and S. Stos-Gale at the Isotrace Lab in Oxford, part of the same RLAHA which was and is the home of Archaeometry; yet how modest the impact of isotope studies in archaeometallurgy has been on the publication profile of *Archaeometry*. There has been a relative surge of isotope-related papers from the 1990s onwards (see below), but the bulk of the discussion and method development appears to have been published elsewhere. There is one remarkable exception to this though, when a paper that was reviewed with contrasting results was openly discussed in the journal. Partly in reaction to the criticism aired by Budd et al. (1993), the Isotrace Laboratory published a series of papers on lead isotope ratios of ores from various regions where they have worked.

Metallography and manufacturing

A major and uniquely metallurgical method to study artefacts is metallography, using optical and scanning electron microscopes / microprobes. The main emphasis of this approach is on the identification of particular structures, preserving some of the manufacturing history of an object. Pioneering work has been done by Gowland (1912) and Bergsoe (1938), followed by the work of CS Smith (collected in Smith 1981). Numerous papers by Tylecote, Lechtman, Kolchin, Bielenin, Pleiner, and Maddin and co-workers during the second half of the last century developed metallography to a routinely applied approach in archaeometallurgy, to mention just a few particularly prolific scholars, representative for a much larger group. Analyzing iron and steel requires accurate determination of the carbon content at levels between 0.01% and 1% by weight. Few of the analytical instruments used in nonferrous metal analysis are capable of doing this, and methods established in industry normally require much larger and better preserved samples than are typically available in archaeology. Here, optical metallography is the most appropriate method not only to determine the carbon content, but to reconstruct the working history of the object under study. Significantly, metallography enables the reconstruction of a sequence of events, as opposed to a mere description of the status quo. Not only is the working history preserved in the microstructure of the metal, but also subsequent changes in composition or condition by use, corrosion or conservation treatment. While much of this, such as grain size and shape, phase identification and detailed composition, can be quantified, it is still the overall and often qualitative assessment of the spatial relationship between different phases and individual metal grains which renders metallography as much an experience-based as a quantitative method. It would go beyond the scope of this review to list the wide range of studies of archaeological metal artefacts which are based on this method, covering all known metals of antiquity, and a number of less well known ones, too, such as platinum (Bergsoe 1938), zinc (Rehren 1996), antimony (Shortland 2002) and aluminium (Bourgarit and Plateau 2007).

Alloying, refining, casting and recycling all produce their own compositionally distinct types of waste material, typically in much lower quantities than smelting and often in close relationship to technical ceramics such as crucibles and hearths. These workshop wastes are different from the primary production residue. Significantly, they are often more removed from equilibrium conditions than most other archaeometallurgical materials. This results in the preservation of intermediate stages of the various operations which were carried out at the workshop, alongside raw materials, intermediate products, and finished products in varying proportions. As in metallography, it is the assessment of the spatial and chronological relationship between the different phases present which enables the microscopist to interpret these residues in a way which is not possible for a more quantitative and instrument-based analysis.

In summary, archaeometallurgy is a rather broad and diverse field, and draws from an equally wide range of scholarly and analytical methods of study. Metals play a fundamental role in the social, economic and technological fabric of almost all postneolithic societies. The study of ancient metal production and manufacturing, and the trade of raw metal and finished metal objects, includes such diverse approaches as optical microscopy, physical, chemical and isotopic analysis, and experimental reconstruction. How is this wealth and diversity of archaeometric approaches reflected in the literature?

Publishing archaeometallurgy

Studies of archaeological metal objects, their production and manufacturing methods were pioneered already in the early 19th century by eminent chemists. M.H. Klaproth (1815) published the first ever quantitative analysis of an alloy, on a Roman coin (Caley 1949). Others include J.F. Gmelin (1786), G. Pearson (1796), J.J. Berzelius (1836/37) and M. Berthelot (1906). Particularly relevant were the works by F. Wibel (1864, 1864) who addressed many archaeometallurgical topics and problems, such as the composition and identification of native copper, more than a century ago. Comparatively systematic studies of the composition of ancient metal objects were performed by v. Fellenberg on bronzes (1866) and v. Bibra on bronzes, iron and silver (1869, 1873). There was little further progress until the late 1920s when the Sumerian Metals Committee was appointed by the Royal Anthropological Institute, triggered by the exceptional finds at the Royal Cemetery at Ur in Mesopotamia (Woolley 1931). It reported on the origin of Sumerian copper assuming that its nickel content could be indicative of the ore source (Desch 1928-1938). From these interim reports it is obvious that the original objective was not really achieved but they resulted in the creation of a further unit, the Ancient Metal Objects Committee in 1939.

In the 1930s a new analytical method was introduced, optical emission spectrometry, nowadays called atomic emission spectrometry. This technique allowed the determination of many elements at trace levels using minute sample masses and was the basis for the development of a new interdisciplinary field, geochemistry. Like their predecessors, geochemist also became interested in ancient metallurgy, and the first programmatic paper on provenance determination appeared in 1934 (Noddack and Noddack 1934). During this time, large analytical programs on ancient metals and ores were started by W. Witter (1935, 1938) and Pittioni (1932) as well as Preuschen

and Pittioni (1937), which lead to the publication of major summary works (Otto and Witter 1952, Pittioni 1957) with a compilation of some 6,000 analyses of prehistoric metal objects, mainly from Europe. This was called the first phase of analytical archaeometallurgy by Härke (1978), who provides a comprehensive summary of the history of archaeometallurgy. In this period the "Ancient Mining and Metallurgy Committee" was founded in London by H.H. Coghlan which initiated analyses of ancient metal artefacts along the lines of the pioneering works of Otto and Witter. The second phase began with another large-scale program, initiated by S. Junghans in Stuttgart: The "Studien zu den Anfängen der Metallurgie" which eventually produced and published more than 22,000 analyses of metal objects (Junghans et al. 1960, 1968, 1974). Many laboratories in Europe (Vienna, Moscow, Baku, Milan, Rennes, London) joined this endeavour with the aim of identifying the composition of metal objects in different periods and identifying the sources from which the raw material came. One of these laboratories was the Research Laboratory for Archaeology and the History of Art in Oxford (Blin-Stoyle 1959, Britton 1961, Britton and Richards 1963). Following the observation that many of the OES analyses were not comparable between laboratories, it was believed that metal analyses by OES were "a waste of time" (Hall 1970). The program was stopped and metal analyses were then performed by neutron activation analysis (Gordus 1967) or by atomic absorption analysis (AAS, e.g., Cowell 1987). This marked the beginning of the third phase according to Härke (1978), characterized by the employment of other and more accurate analytical techniques for metal analysis than OES.

The work focused on metal artefacts was complemented by studies of the primary production. While chemists pioneered the analysis of metal objects, it is mostly metallurgists and geologists who drove the development of the metallurgical aspects of archaeometallurgy. General works on the history of metals and metal production appeared from the second half of the 19th century onwards (Zippe 1857, Rossignol 1863, Andree 1884, Rössing 1901, Neumann 1904, Gowland 1912; Bergsoe 1938; Marechal 1962). These were accompanied and followed by papers by, e.g., Morton and Wingrow in the late 1960s and early 1970s, CS Smith from the 1950s to the late 1970s, R. Tylecote from the 1960s to the 1980s, H.G. Bachmann from the 1960s and R. Maddin from the mid 1970s onwards, often in collaboration with mining archaeologists such as B. Rothenberg, G. Weisgerber, B. Jovanovic and C. Domergue. The literature here is vast, and dispersed over a wide range of journals in the engineering and natural sciences, archaeological journals, and excavation monographs. Good bibliographies are contained in books such as Tylecote (1987), Rostoker and Bronson (1990), Craddock (1995) and Pleiner (2000).

Significantly, this rich history of serious and often large-scale studies has been published predominantly in form of monographs, or as articles in established journals in the 'mother disciplines' of the authors. Until 1958, there was no specific publication outlet dedicated to the interdisciplinary work crucial for the new progress made.

Specialist journals

These pioneers in archaeometallurgy literally had to invent the field, and had little pre-existing academic structure to work with. This lack of structure included the absence of dedicated journals, and as a result they founded their own journals: in 1966 Ronald Tylecote established the *Journal of the Historical Metallurgy Society* (now

known as Historical Metallurgy). It appears that the very term archaeometallurgy was coined only in 1973 by B. Rothenberg (Goodway 1992), when he established the Institute for Archaeo-Metallurgical Studies, publishing much of its work in the *iams* newsletter, now the iams journal. Significant work in archaeometallurgy grew out of the work of the scientific laboratories of major museums, such as the British Museum, the Deutsches Bergbau-Museum, and the collective Berlin museums. Both the BM and the DBM organised series of international conferences in archaeometallurgy, providing important venues for the exchange of ideas and development of projects; many of these conferences were published, either as British Museum Occasional Papers, or as supplements ('Beihefte') to the DBM's house journal, Der Anschnitt. A series of conferences under the title Beginnings of the Use of Metals and Alloys, BUMA, was established by R. Maddin in the mid 1980s, and recently had its sixth incarnation in Beijing. The Bulletin of the Metal Museum was set up in 1976, the year after the foundation of the Metals Museum by the Japan Institute of Metals. It was a special case in that it did not so much publish primarily research by the staff or members of the backing institution, but relied heavily on invited papers and submissions from outside the museum. This journal ceased to exist in 2003, due to the closure of the Metals Museum. Archeomaterials, starting in 1986, was explicitly more wide-ranging than focusing on ancient metals, although many of its papers were concerned with archaeometallurgy; in contrast to the other examples mentioned before, it had no institutional or organisational structure behind it but was backed by one individual, William Rostoker. Following his death in 1991 Archeomaterials ceased to appear in 1993, after only seven issues.

Three journals set up in the tradition of *Archaeometry* need to be mentioned. The *Journal of Archaeological Science* first appeared in 1974, followed by *Revue d'Archeometrie*, in 1977; both cover the entire range of archaeometry, including papers on archaeometallurgy, without any particular link to a laboratory. The annual *Berliner Beiträge zur Archäometrie* was first published in 1975 by the Rathgen Research Laboratory, founded in 1888 as the Chemical Laboratory of the Royal Museums in Berlin. Like *Archaeometry*, it is closely linked to a particular laboratory; it carried a range of papers on archaeometallurgy, most notably reports on the composition of metal artefacts, conducted in the tradition of the earlier large series of object analyses.

Archaeometallurgy in Archaeometry

Thus, and while there was and is a range of specifically archaeometallurgical journals and series, *Archaeometry*, starting in 1958, was ahead of the game by a decade or two. Its position and starting vision were typical of the time; as stated by Edward Hall, its founding editor, it was meant to report (primarily but not exclusively) on work done by staff of the Research Laboratory for Archaeology and the History of Art at Oxford University. It aimed to rapidly circulate results of completed research as well as to report on only partially successful work 'not worthy of normal publication', and of interim results of work in progress (Hall 1958: Foreword).

This policy of also publishing interim and unsuccessful work did not prevail for long. Soon, *Archaeometry* became an outlet for full-blown research papers, with the same standards of peer review and subsequent delay in publication as other major journals. However, the emphasis on work done within the laboratory and its various cooperations was less easy to overcome, and remained visible for decades to come. This

is not the place for a detailed breakdown of papers by sub-discipline within the journal, and how this changed over time; suffice it to say that in the first decade, papers on dating methods and geophysical prospection featured both equally strongly, with the latter very abruptly disappearing after 1971, and the former gaining significantly from 1970 onwards. There are volumes in the 1970s where papers on dating make up between one third and one half of all the published papers. From the 1980s onwards things become more balanced, with relatively stable ratios between papers on dating, ceramics, organic materials, metals, and other topics.

Looking more closely at the number and topics of papers in *Archaeometry*, archaeometallurgy started strong with two out of five in the first year. But while the number of papers published quickly increased to a typical 15 to 20 per year throughout the 1960s, contributions on metals remained at around 1 to 3 papers per year (ppy) until 1971. By this time, the average number of papers per year in the journal overall had risen to more then 20, and remained at 20 to 25 ppy for the next 25 years. Throughout this period, archaeometallurgy contributed regularly between 2 and 5 ppy; a figure which has not significantly improved since then, hovering at 5 to 6 ppy for the last ten years or so. At the same time, the overall number of papers in the journal has increased significantly. From 1995 onwards there were around 30 ppy overall, which further increased to 40 to 50 ppy in the last five years. Thus, we see a strong and sustained increase in papers overall in Archaeometry, particularly for the last decade or so, but less so in papers dealing with archaeometallurgical topics. Based on pure numbers, archaeometallurgy papers were most prominent in the mid-1970s when they constituted around 20% of all papers published; since then, the share of archaeometallurgy papers within Archaeometry has fallen to nearer 15%, and even as low as 10%.

Despite the broadening of the scope and range of topics published in Archaeometry beyond the immediate interests of the staff and associates of the RLAHA, there is still a rather specific profile of archaeometallurgy visible; some topics are strongly represented in Archaeometry, while others are nearly absent. Most notably, there is a very strong focus on chemical analysis of gold and silver coins and bronze artefacts; the entire second volume is on bronze analysis. Coin and artefact analysis were almost the only archaeometallurgical topics for the first five years of publication, and up to the mid 1980s there were also regularly one or two papers on the development of methods of chemical analysis of metals, often using coins as test cases (e.g., Meyers 1969). Thus, coins feature both as objects of study in their own right, and as convenient (and relevant) test materials. From the early 1970s on they are increasingly accompanied by papers reporting the composition of other types of metal artefacts; most often bronze objects, and discussions of analytical method developments (e.g., Hughes et al. 1976). These three subgroups (coins, bronze artefacts, method development) make up, to almost equal parts, the bulk of all archaeometallurgy papers for more than a decade, from 1972 to 1985.

However, the frequency of 'coin papers' drops dramatically in the mid 1980s: before 1986 coins were typically represented with one or 2 ppy, but from 1986 onwards this dropped to an average of 0.5 ppy. This decline is initially not compensated for by the publication of other archaeometallurgy papers, and there is a noticeable lull in archaeometallurgy papers in *Archaeometry* from 1986 to 1991. Not only had the supply of coin papers dried up, but also the publication of papers on other metal artefacts has all but ceased. Thus, for five years there are only 2-3 ppy on archaeometallurgy.

A major change happens in 1992, when for the next twelve years this trend is reversed by the sudden and sustained emergence of isotope-related papers. However, this is rather late, with the first paper on lead isotopes in 1985 by Mabuchi et al., twenty years after Brill and Wampler (1965) introduced the method to archaeology. The next appears in 1988, and it is not before 1992 that a constant delivery of two or three or so papers per year sets in. This had been briefly commented already above in the context of lead isotope studies in archaeometallurgy. Due to the importance of LI analyses for the reconstruction of ancient trade pattern it is of major interest and direct significance for archaeology, probably more so than the more difficult to interpret trace element patterns of objects.

Most remarkable is the very low frequency of papers to do with iron metallurgy. This is a massive and significant deviation from the archaeological reality on the ground, and the quantity of excellent work done in the field. Iron is of overwhelming importance in nearly all cultures of the last two millennia or so, from the Roman period onwards in Europe, from the Han dynasty onwards in China, and throughout the metal-using history of sub-Saharan Africa. Not only are studies of iron artefacts or metallography in general exceedingly rare in *Archaeometry* (Knox 1963 and Charles 1973 are rare exception), but also studies in iron smelting. Some of this may be the mirror effect of the prevalence of iron-related papers in *Historical Metallurgy* with its strong tradition of iron- and steel-specific papers. Only in the last few years do we see more iron-themed papers published in *Archaeometry*, probably indicating a broadening of the author base of the journal.

A similar *lacuna* is the near-total absence of papers presenting or discussing primary production or more technological process-oriented studies. The recent papers on early copper production in the Alps (Höppner et al. 2005; Tumiati et al. 2005) and on reconstructed EBA copper smelting (Pryce et al. 2007) are probably the first papers concerned with copper smelting ever to be published in *Archaeometry*, and Heimann et al. (2001) and Paynter (2006) the first ever for iron smelting.

Thus, in summary it is fair to state that *Archaeometry* has been a consistent, but never a major outlet for archaeometallurgy. Over the half century of its existence, about 185 archaeometallurgy papers have appeared, many of them innovative and stimulating further research. Remarkable is the rather clear profile of most of these, giving archaeometallurgy in *Archaeometry* its very own flavour.

Conclusion

Archaeometry is turning 50 this year. During this time it has grown out of its origins as an outlet for some ongoing work from a particular laboratory, into a leading journal covering the entire range of archaeometric research, with authors from around the globe. Archaeometallurgy as one of the constituting parts of science-based archaeology has a firm place within the journal, but with a particular profile. The range of archaeometallurgical papers typically printed in Archaeometry differs significantly from the overall range of activity in the field. This reflects partly the much earlier origins of archaeometallurgy, particularly in central Europe, which had established their own traditions to publish in other journals, but also the emergence of a number of smaller, specifically archaeometallurgical journals soon after the Archaeometry first appeared, which have their own focus and emphasis, and cater for a considerable amount of work done in archaeometallurgy. Thus, more process-

specific work, and work concerned with iron making and iron or steel objects, is often published in either *Historical Metallurgy* or in *Archeomaterials* (while it still existed); papers with particular reference to Asian metallurgy often appeared in the *Bulletin of the Metals Museum*, printed in Japan. *Archaeometry* in contrast traditionally has an emphasis on physical and chemical approaches to archaeometry, manifest in the majority of papers dealing with chemical analysis of coins and other metal artefacts, often combined with method development, whereas papers based on metallography or the analysis of waste materials such as slags and technical ceramics are much underrepresented. Of particular interest is the situation concerning the application of lead isotope studies in archaeometallurgy. The Isotrace Lab, as part of the RLAHA, has been for many years one of the two leading laboratories in this area, but relatively little of this shows in the journal *Archaeometry*. More recently, this division is blurring, and the last five years have seen somewhat increasing numbers of papers on both iron, and on metal smelting in general, which were virtually absent from the first 40 years of publication activity.

It would be an altogether different (but certainly interesting) paper to investigate the conscious and subconscious decisions taken, by authors and editors alike, which drive this pattern in publication behaviour. Suffice it here to say that these pattern have their roots in the past, shape the present, and will certainly continue to exist in one form or another in the future. We are confident that *Archaeometry* will further develop its special contribution to archaeometallurgy, and we wish it every success in this, for decades to come!

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Figure 1: The archaeometallurgy cycle. After Ottaway 1994.

