# METALLURGY AND METALLOGRAPHY
# IN NUMISMATICS

## Introduction

Numismatics concerns itself mainly with coins as documents. But the study of coins as artefacts can make an important contribution by elucidating both the techniques by which they were made and their metallic composition.

Ancient metal-working techniques have intrinsic interest, but for numismatist the aim of examining mint procedures is to establish chronology and attribution through similarities or divergences in minting technique. The likely productivity of each die, and hence the total size of the issue which could have been produced from the number of dies actually observed, can also be estimated by considering the practice of the mint.

Systematic differences in composition, too, can be used to determine chronology and mint attribution, as well as the sources of raw material. But it is changes in fineness, especially debasement, which have been the central concern of most analytical work. Once these are as clearly known as possible, we can approach an understanding of the intentions of the moneyers, and finally, perhaps, of the minting authority.

A considerable literature now exists on both minting technique and composition.[1] There has, however, been a tendency to treat the topics separately, and to under-estimate the subsequent effects of various environments, such as wear, corrosion and cleaning. But metallurgy, especially through the metallographical examination of coins, reveals the complex inter-action and inter-relation of all these factors. I present here a systematic, but non-technical, account of the effects of each stage in the minting process, and later changes, on observed composition and structure.

## The raw material

The first step in minting must be the collection and preparation of material. Information on the sources of the material used for the coins, and the methods by which it was prepared for use, can be derived from their composition.

[1] See W.A. Oddy (ed.) Scientific techniques in numismatics, M.J. Price, E.M. Besly, D.W. MacDowall, M.E.P. Jones, W.A. Oddy (eds.) *A Survey of numismatic research 1987-1983*, International Numismatic Commission, 1984.

The ratio of the four lead isotopes, $Pb^{204}$, $Pb^{206}$, $Pb^{207}$ and $Pb^{208}$, unique to the lead in each silver ore is particularly valuable for the study of provenance, since it is not affected by extraction, refining, coin fabrication or corrosion. In certain early coinages, such as the earliest Greek silver, lead isotope ratios in the coins can be matched to those in known ores, such as those of Laurion and Siphnos[2]. But as soon as any appreciable mixing of silver stocks takes place, in this region probably as early as the transfer of the treasury of the Delian League to Athens in 454 BC, the method becomes useless. Even in later periods, however, it may be possible to suggest that raw material came from certain areas: for instance the nickel present in eastern copper ores may explain why mid-third century radiates minted at Antioch have nearly double the proportion of nickel found in contemporary issues at Rome.[3] Re-used coin was also an important raw material: the presence of 1.5% zinc in a dupondius of Vespasian (RIC 90) analysed by Lawrence Cope was attributed by him to a mixture of silver with coinage brass.[4] It has also been argued that the higher proportion of tin and lead in early radiates of Claudius Gothicus issued at Milan, compared with Rome, is due to the use of larger quantities of sesterces in the melt.[5]

Gold and silver in ancient coins were normally prepared and purified by cupellation, or preferential oxidation of metals lower down the chemical series. The greater the proportions of silver and lead (in gold) or gold and lead (in silver), the less the efficiency with which the operation has been carried out. In debased silver coinages, however, some of the gold and lead will be attributable to the base mixture to which the silver was added. The proportion of silver is naturally carefully controlled, in most cases, but there a few exceptions. It has been shown that the silver content of a group of tetradrachms probably

[2]) N.H. GALE, W. GENTNER and G.A. WAGNER: Mineralogical and geographical silver sources of archaic Greek coinage, in E.T. HALL and W.A. ODDY (eds.): *Metallurgy in Numismatics I*, 1980, pp. 3-50, (hereafter *Metallurgy*, 1980). See also R.H. BRILL and W.R. SHIELDS: Lead Isotopes in Ancient Coins, in E.T. HALL and D.M. METCALF (eds.): *Methods of Chemical and Metallurgical Investigation of Ancient Coinage*, 1972, pp. 274-305, (hereafter *Methods*, 1972).

[3]) L.H. COPE: The metallurgical analysis of Roman imperial silver and aes coinage, *Methods*, 1972, pp. 44-5.

[4]) Ibid. p. 45.

[5]) L.H. COPE: The nadir of the imperial antoninianus in the reign of Claudius Gothicus 268-270 AD, *Numismatic Chronicle* 1969, pp. 145-61. See also C. BRENOT, H. HUVELIN and J.-N. BARRANDON: Le metal des antoniniani de Claude II: un aspect des rapports entre l'atelier central de Rome et les ateliers de Milan et de Siscia, *Atti del Convegno Internazionale di Studio, Milano 1983*, 1984, pp. 173-81.

minted at Rome under Severus Alexander was kept extremely close to that of Alexandrian examples, yet contemporary denarii show a much wider variation.[4]).

## The melt

Melting probably took place in a fixed-draught charcoal-burning furnace, which could reach a maximum temperature of about 1600 °C. This has consequences for the interpretation of observed composition and of its relation to the intended composition. Certain metals, (such as calcium, titanium and aluminium) cannot be reduced below this temperature, so their reported presence in some analyses[7]) is probably due to contamination from earth, slag or cleaning solutions. Some metals which can be reduced, (such as selenium) have not been found, perhaps because they escaped as vapour. Conversely, others (such as platinium and ruthenium) could not be oxidized or removed below 1600 °C, so that once introduced in to a stock of coinage metal, they will always persist as markers of provenance. The sharp rise in platinum content in Roman gold coinage from 317, and especially between 346 and 368, suggests that that new sources of gold were being used[8]). A number of laws in the Theodosian Code controlling production and reserving part for the state suggest that new mines were being exploited at this period. Another source may have been the temple treasures confiscated by Constantine.

But there remains a largely unquantifiable gap between the composition of the whole melt and that of an individual coin. The heavier metals, especially lead, tend to sink towards the bottom of the vat. Others, with an affinity for each other, such as copper and tin, or silver and lead, tend to draw together. Thorough stirring and mixing will keep the melt fairly homogenous, but if this is carelessly done, a proportion of coins, at least, will not have the intended composition. The importance of these effects is shown by an experiment carried out by C. T. Peters on a melt of highly-leaded argentiferous bronze with a similar composition to the folles minted at Arles under Constantine. With

[4]) A. BURNETT and P. CRADDOCK: Rome and Alexandria: The minting of Egyptian tetradrachms under Severus Alexander, *American Numismatic Society Museum Notes* 28, 1983, pp. 109-118.

[7]) E.G. G.F. CARTER: Preparation of ancient coins for accurate X-ray analysis, *Archaeometry* VII, 1964, pp. 106-113.

[8]) C. MORRIS, C. BRENOT, J.-P. CALLU, J.-N. BARRANDON, J. POIRIER, R. HALLEUX: *L'or monnayé I: purification et alterations de Rome è Byzance*, Cahiers Ernest-Babelon 2, Editions de CNRS 1985.

a melt containing in all 1.39% silver, the silver in a cast strip poured from it increased from 1.15% at the start to 1.57% at the end. The proportion of lead, 12.50% in the melt, increased from 10.35% to 14.62%[9]).

The evaporation of certain metals, especially zinc, with a melting point of only 906 °C, may also make the final product diverge from the intended composition. It has been estimated that the resulting enrichment in silver may be about 4% when the process is closely controlled, and up to 10% when it is poorly controlled[10]).

## Blanks

Coin blanks can be produced from a melt in two basic ways. The metal may be poured out and allowed to cool in order to form a cast strip or sheet from which flans of the right size and shape can be cut, or the metal may be poured out in droplets, which can be beaten out when cool. Reworking of the cast buttons or sheet may require reheating (or annealing).

Study of the macrostructure of the coin, as revealed in a section, may show which process was used, and with what modifications in detail, in particular through the distribution of gas cavities or oxide inclusions. The medieval English penny, for instance, underwent a change from one technique to the other in the late thirteenth century as has been shown by microphotographs of oxide inclusions in the section. The Long Cross penny, issued from c. 973 to 1279, displays mainly horizontal inclusions, but at the edge the inclusions are vertical, as expected for a flan punched from a cast sheet of metal. Its successor, the penny of Edward I, shows no trace of vertical inclusions, which suggests that the flan was produced by beating out a cast button[11]).

Here the change in technique coincided with a change in type, but this is by no means always true: X-ray examination has shown that some blanks, at least, of coins in the Roman series, were cast on edge, and display a segregation of lead towards the lower edge, like that described above in a melt[12]). Others

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[9]) C.T. PETERS: *A metallurgical investigation of the 1/96 libra Arelate folles of the Constantinian era*, unpublished M.Sc thesis, University of Surrey, 1970.

[10]) L.H. COPE: *The metallurgical development of the Roman imperial coinage during the first five centuries AD*, unpublished Ph.d thesis, Liverpool Polytechnic, 1973, p. 125 and table X.

[11]) D. SELLWOOD: Alterations in mint technology for the Edwardian penny, *Metallurgy*, 1980, pp. 178-180.

[12]) L.H. COPE: The metallurgical analysis of Roman imperial silver and aes coinage, *Methods*, 1972, p. 13 and plate IV, 8, for a sestertius of Septimius Severus.

were formed from cast buttons, like the Edwardian penny. But this does not seem to be linked to stylistic or compositional changes.

Further confirmation and amplification of these effects, together with their consequences for the composition of coins, can be provided by the study of microstructure, which will be discussed below.

## Minting

Distinguishing between pieces minted by casting or striking can be very simple: the tiny bubbles and slight lack of definition which characterize a cast piece can usually be detected by the naked eye, or at least with an ordinary magnifying glass. Cast pieces may also fail to ring in the way characteristic of struck pieces. In the Roman series a cast coin is, of course, a forgery (whether ancient or modern). Tooling, another indication of a fake, can also often be seen with the naked eye.

More sophisticated examination, using the technique of metallography, is needed to determine further details of the technique, such as whether the blank or coin has been worked when hot or cold. A section is prepared, which is then highly polished and etched with acid, before being examined under a metallographic microscope, and possibly even by X-ray diffraction.

## The structure of metals

Understanding the results requires an understanding of the basic structure of metals[13]). All metals consist of a three-dimensional jigsaw puzzle of crystals, also called grains. Within each grain the atoms are regularly arranged, so that for instance, in one form of brass, each zinc atom has eight copper neighbours, and each copper atom has eight zinc neighbours. The form of the crystal or grain is determined by the conditions under which it was created. When heated, the atoms re-arrange themselves towards the most stable array at that temperature, and at each successive higher temperature. If cooling then occurs slowly, the process reverses itself, but if the metal or alloy is cooled suddenly, the atoms have no time to re-arrange themselves and their kinetic energy is very much reduced, so that they are left in an array which is unstable at that temperature. Annealing by reheating allows transformation towards stability to take place.

[13]) For this and the following section, see further A. COTTRELL: *An introduction to metallurgy* (2nd edition), 1975.

### The structure of alloys

The structure and transformations of alloys are much more complicated. A few alloys, such as cupro-nickel, have only one type of grain present throughout their range of composition. In other words, they are always single-phase alloys. But most alloys have two or more types of grain (two phases) at certain ranges of composition. These are usually identified by Greek letters. For instance, in brass containing up to 30% zinc, only one type of grain is present (α). At about 35% zinc a new type of grain appears (β), and at about 45% there are equal numbers of the two types of grains, α and β. Above 50% all the grains are of the new type. Thus brass is a two phase alloy between 35% and 50% zinc, and single phase above and below these figures.

This assumes constant temperature: for any given composition different phases, or combinations of phases, will be present, depending on temperature.

Phase diagrams, showing what phase or phases, should be present for any combination of composition and temperature, have been worked out for most binary alloys, many ternary alloys, and a few four- (or more) element alloys. They only describe the alloy in equilibrium: when it has been left in an instable condition due to quenching or annealing, as explained above, other combinations of phases may be present, providing valuable evidence of the mechanical and thermal history of the artefact. For instance the absence of the brittle δ-phase in bronze, which would normally be formed during cooling, indicates that the metal has undergone prolonged annealing. Naturally, however, phase diagrams have been prepared mainly for alloys now in commercial use, and very little work has been done on the more complex alloys found in ancient coinage.

### Minting resumed

Now that the basic terms and concepts have been defined, we can return to the discussion of the details of minting techniques and its effects on the internal structure of the coin, as revealed by the metallographic study of the coin section.

To deal first with structure, the most important effect of phase formation in coinage alloys is micro-segregation within the blank or coin. This is on a far smaller scale than the segregation by density already mentioned. It is due essentially to differences between the liquid and solid solubilities of the phases.

The most serious segregations are caused by metals which are virtually insoluble in the solid state, and thus form phases of almost pure metal, such as lead, iron and cobalt. Unfortunately these are also the most common impurities in copper-based coins.

Partially soluble elements, such as zinc, tin and silver, cause less serious problems, but separate phases consisting largely of these elements will tend to form.

The differing freezing points of these phases, once formed, may cause segregation by a number of different mechanisms. In the simplest, known as coring, the phase with the lowest freezing point migrates through the alloy in the direction of solidification, so that it ends concentrated in the centre of the cast. In another form, dendrites, or tree-like solid growths may be created by a phase with a higher freezing point than the surrounding liquid, and may form segregated cells. Inverse segregation, the opposite of coring, which leads to beads of a still liquid phase appearing on the surface of a cast, may be caused by the formation of interdendritic channels along which the liquid is sucked.

Turning now to evidence of technique, cold working in general deforms the grains, while hot working reforms them. Thus tooling can often be detected by irregularities and cracks in the grain stucture, even if it has not been made obvious by the corrosion which they encourage. The sudden chilling caused by cold dies striking a warm blank may also cause cracks.

Hot working, especially annealing, which is used to make blanks less brittle, is characterized by large grain size, and in particular by the presence of annealing twins. These are twin crystals, sharing one face, which form during the re-growth and re-formation of crystals as the temperature is raised. Quenching fixes this arrangement by lowering the temperature very sharply, and also restores the hardness lost by annealing.

Hot or cold striking has important consequences for die productivity, as an experiment by David Sellwood has shown. He found that 8000 replica Greek tetradrachms could be struck from one reverse die, and 10,000 to 16,000 from one obverse die, if heated blanks were used, but only 4,000 and 8,000 respectively if the blanks were cold[14]).

There are also important surface effects, arising from the technique of minting. Lead may be concentrated near the surface as a result of annealing, but for the Roman series, the most important effect is the surface enrichment in silver, which occurs even in the very debased silver issues of the third century. It has been suggested that the remarkably silvery appearance of these radiates was due to the quenching of the hot-worked blanks in vinegar or acid fruit juice[15]). The practice was probably begun much earlier simply to remove the

14) D. SELLWOOD: Some experiments in Greek minting techniques, *Numismatic Chronicle* III, 1963, pp. 217-231.

15) L.H. COPE: Surface-silvered ancient coins, *Methods*, 1972, pp. 267-8 with fig. 1 and plate XIX, no. 1.

oxide layer, formed during annealing. Of the separate copper-rich and silver-rich phases present, the copper-rich phase was preferentially attacked by the acid, leaving mainly silver exposed at the surface. Further beating of the blank before striking created a silver skin of similar thickness to the silver-rich phase in the body of the coin, and continuous with it.

But below about 8% silver, the silver-rich phase would no longer be continuous, and another explanation must be sought for the silvery appearance of the better-preserved coins. Lawrence Cope found by experiment that two methods were possible, though of course their actual use cannot be proved. In the first, for flans containing 5-8% silver, cold hammering followed by annealing led to the exudation of lead, together with silver, on the surface of the coin. Quenching in water or dilute acid then leaves a blanched silvery surface. For flans containing less than 5% silver, it was necessary to immerse the blanks in a bath of molten silver chloride. Since the chlorides of copper, lead and tin are more stable than that of silver, pure silver would tend to be displaced and to form a wash on the surface on the coin[16].

Brasses and bronzes are also affected by these phenomena, but to a lesser extent. Hot-worked brasses are prone to dezincification of the surface, and surface enrichment in tin may occur in some bronzes as a result of annealing.

*Changes after minting*

Once minted, coins in circulation suffer many changes: they may be broken, filed, pierced or simply worn down. All of these complicate interpretation of their composition, principally by reducing the total weight of the piece by an amount which it is difficult to quantify. An estimate can at least be made for rates of loss throught wear, based on modern figures. French gold coins of the nineteenth century lost 1-2 mg each year, while silver coins lost 10 mg, probably due to their more active circulation[17]. Lower value coins would usually circulate still more quickly, but the greater hardness of alloys would tend to slow down the resulting loss of weight. But it is important to note that the weight loss applies regardless of denomination, so that the smaller the coin, the greater the percentage loss of weight in a given period. The question is complicated still further by the wide range of weights in some ancient issues, by varying rates of hoarding and circulation, and by the effects of corrosion[18].

[16]) Ibid. p. 275.

[17]) D.M. METCALF: The application of statistics to numismatics, *PACT* 5, 1981, p. 11.

[18]) P. GRIERSON: Coin wear and the frequency table, *Numismatic Chronicle* III, 1963, pp. i-xvi.

Museum specimens, at least, may also tend to be heavy and unworn examples from the issue.

The main cause of actual change in their composition and structure is, however, corrosion. This affects both the total composition of the piece, and the composition of its surface, which is especially important for many forms of analysis. I will deal first with the total composition.

There are three main mechanisms, of which oxidation is the most important for museum specimens, and electro-chemical and acid corrosion are most important for archaeological finds.

Electro-chemical corrosion may occur through the formation of a differential aeration cell or by galvanic action. In the first, positive metallic ions are formed by solution of the metal at the anode, while the electrons liberated combine with dissolved oxygen to form negative hydroxyl ions at the cathode. The resulting metal compound is deposited between the anode and the cathode, while the anodic area is deeply corroded. Thus metals below hydrogen in the electrochemical series are corroded readily, those above, such as copper, silver and gold, less so. The process requires the presence of some air, since dissolved oxygen is necessary for formation of the negative ions, and is accelerated by the greater conductivity of salt solutions, such as seawater.

A more serious form of corrosion may occur when different metals are contiguous, whether in a hoard containing both silver and bronze, or on a microscopic scale, in different phases. Galvanic action also works by the creation of an electrical cell, but in general the less noble metal (rather than the submerged part of the artefact) will become the anode and be deeply attacked, though the protective action of oxide films may complicate this.

Corrosion by acids, such as those present in many types of groundwater, is the most damaging of all. The metal is dissolved into positive ions, but there is enough hydrogen in the acid to neutralise the resulting electrons, so no cell is formed. The reaction simply continues to remove metal as long as there is hydrogen present, though oxide films offer some degree of protection here also.

As a result of these processes, Cope suggested that anaerobic conditions are far more unfavourable to preservation of coins in soil than aerobic conditions: within a period of 16 years corrosion of lead and zinc was five to ten times faster in the absence of air[19]. No work has been done on the effects over

[19] L.H. COPE: *The metallurgical development of the Roman imperial coinage during the first five centuries AD*, unpublished Ph.D thesis, Liverpool Polytechnic, 1973, p. 72 and fig. 3.

archaeological time, but it seems likely that a plateau, or state of equilibrium would be reached fairly quickly.

Oxidation, by contrast, which occurs through reaction with oxygen in the air, may be the most benign form of corrosion: a film or skin of oxide is quickly formed which protects the metal from further damage. This patina is easily removed and need not affect the results of analysis at all.

But the effects of oxidation on coinage alloys may be devastating, depending on their degree of heterogeneity. The volume of a metal oxide is always higher, sometimes much higher, than that of the unaltered metal. As a result part of an alloy will tend to be forced towards the surface of a coin, and holes may form. In a single phase alloy one constituent may be more easily affected, and in a polyphase alloy one phase may be more attacked than others.

Copper-silver alloys are by far the most seriously attacked, since they are polyphase alloys, in which the copper-rich phase is far more vulnerable to oxidation and forms an oxide with nearly two-thirds greater volume than the metal. As a result the recorded percentage of silver by weight may be greatly increased, for instance from 71% in the unaltered part of a coin to 81% in the corroded part, in analyses of denarii carried out by Condamin and Picon[20]). The effect is naturally greater in more debased silver: one of these coins, containing 45% silver in the unaffected core, had 58% in the corroded part. A tenth of the ninety denarii used in these experiments were at least 80% oxidized by volume, and «many more» (we are not unfortunately told how many, or to what degree) were less seriously affected. It is even more unfortunate that no indication of the source of these coins is given, though it is likely that they were museum specimens. Archaeological finds might well be much more seriously damaged.

Alloys containing more than a trace of lead are also very vulnerable to corrosion, for similar reasons. Lead is, as I have already mentioned, almost immiscible in the solid state with other metals, and forms phases which are almost entirely lead. As lead corrodes very easily, the lead phase, dispersed throughout the substructure, may provide a pathway for corrosion deep in the coin. This generally results in a lowering of the observed proportion of lead. Some 4%-5% of 'about 250' coins in Condamin and Picon's study were affected[21]).

[20]) J. CONDAMIN and M. PICON: Changes suffered by coins in the course of time and the influence of these on the results of different methods of analysis, *Methods*, 1972, pp. 51-2.
[21]) Ibid. p. 53.

Copper-tin alloys suffer extremely complex and variable alterations, depending on the number and type of phases present, and the presence or absence of dendrites, which in turn depend on the fabrication route for the coin, as described above. They are most likely to be affected if there are several phases present, but again, according to Condamin and Picon, only 4%-5% of 'about 200' coins they examined had suffered any corrosion. The proportion of tin is likely to be increased[22]).

Copper-zinc alloys, or brasses, are least affected by corrosion, since, as explained above, they are single phase and homogeneous. Corrosion is most likely to occur as a result of the stresses set up by cold-working, and to result in a decrease in the proportion of zinc observed.

These phenomena are further complicated by surface effects, which are mainly caused by the redeposit of a metal or metal oxide. Silver is frequently redeposited after being displaced by copper oxide, which may give a misleadingly bright surface to a deeply corroded coin. Tin oxides tend to stay on the surface, while zinc compounds are easily removed. Lead and its compounds are seldom found on the surface as a result of corrosion. Most of these phenomena can be prevented from affecting even superficial methods of analysis by scouring the part of the coin to be analysed to a depth of 0.2mm, but only if the alloy is homogeneous.

Vulnerability to corrosion is increased by stresses in the metal set up by hot- and especially cold-working, or by poor gating of the cast. But a uniform cast surface will always be better preserved than any struck or worked surface.

Finally, the modern processes of collecting and conservation may also complicate the interpretation of composition and structure. As already mentioned, coins in museums will tend, on the whole, to be the finest and heaviest examples of their types, thought those coins given up for the older destructive types of analysis are likely to have been poor, corroded specimens. Conservation of coins, both from museums and archaeological sites, may pose greater problems, especially when conservation records are incomplete or missing. Cleaning may remove traces of silvering or blanching, or increase the degree of surface enrichment caused by segregation or corrosion. Traces of silver may have been added by cleaning in silver nitrate solution, while lead may be removed from bronze coins by prolonged washing. Electro-chemical reduction may leave traces of metals from the solution on the coins, or fail to remove metals

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[22]) Ibid. p. 53.

deriving from the original container or the soil[25]). The removal of corrosion products may in itself, as shown above, falsify interpretation of coin composition.

### Conclusion

This account is a preliminary, and inevitably selective, survey of a large and complicated subject. But I hope that it will guide non-specialists in the evaluation and interpretation of earlier work, and encourage greater integration in the study of minting technique and coin composition.

[25]) W.A. ODDY: Conservation and the requirements of the scientist in numismatics, *Numismatics and Conservation*, Occasional Paper No. 1, Department of Archaeology, University of Durham, 1980, pp. 35-36.