

Microstructural and Compositional Characterization of Roman Coins[†]

I. Calliari,^{1*} M. Magrini,¹ A. Zambon,¹ P. Guerriero² and R. Martini³

¹ Department of Mechanical and Management Innovation, University of Padua, Padua, Italy

² Italian National Research Council (CNR), Padua, Italy

³ Numismatic Museum, Castello Sforzesco, Milan, Italy

Scanning electron microscopy (SEM) with energy-dispersive microanalysis (EDS) and energy-dispersive x-ray spectrometry (EDXRS) were used to investigate the composition and microstructure of some Roman coins from the Julius Caesar and Octavianus periods. The samples were Cu-based alloys with different surface conditions. The microstructural examination supported the chemical analysis data, explaining compositional changes from the surface to the bulk. Compositional data confirm the numismatic interpretation: coins issued by Julius Caesar were orichalcum (Cu–Zn alloy) coins whereas asses issued by Octavianus were Cu–Pb–Sn coins. Copyright © 1999 John Wiley & Sons, Ltd.

INTRODUCTION

This research is a part of a project aimed at supporting numismatic classification with chemical and physical coin examinations. Chemical and metallurgical analysis are powerful tools in numismatic research,^{1,2} as the composition and microstructure of ancient coins can provide information on the alloy employed, melting technology, mints, dating, relative metallic values and remelting of previously struck coins. They also allow one to correlate variations in alloy composition with debasements and to distinguish imitations from authentic specimens.

As regards Roman coinage, even though many names are used to describe copper alloys in the numismatic literature,¹ the Romans differentiated well between the copper alloys they used. Most of the alloys are varieties of *aes*, which is usually translated as 'bronze,' although it also has the meaning 'copper alloy.' 'Aurichalcum' (like gold) or 'orichalcum' was the specific term employed for brass.

Several workers^{3,4} have described the technical and political characteristics of Roman orichalcum coinage, pointing out the brilliant policies of Augustus for *aes* coinage in which the primary mints of Rome and Lugdunum struck in pure copper; whereas the mint of Antioch struck in bronze² and all the others were apparently directed to avoid this primary composition. As zinc was state controlled, estimates of the relative

value of zinc and copper in different coins and periods were also performed. Supposing that the value of orichalcum coins depends on their total weight and on the percentage of zinc they contained, probably the Romans had a different standard for the orichalcum content of each denomination of coin struck under different emperors. The value of zinc added to coins must have been tariffed at some given figure for all denominations; during the reign of the Emperor Nero, for example, 1 g of zinc had the same value as 6 g of copper.⁵

In this study the results of the analysis of the following groups of coins are reported: 13 republican asses issued by Octavianus; 12 Julius Caesar orichalcum coins; 1 Claudian sesterce; 1 Augustan sesterce; and 1 Sextus Pompeius as.

The aims of the research were as follows: to correlate the chemical composition obtained with EDXRS⁶ and EDS with the alloy microstructure; to determine the usefulness and the limitations of surface, non-destructive and easy-to-use investigation techniques in numismatic research for analysing large quantities of coins, including materials in public collections that cannot be subjected to any surface modification;⁷ and to correlate the numismatic hypothesis with chemical results.

EXPERIMENTAL

Thirteen republican asses issued by Octavianus

The republican asses, known as the *Divos Julius* series,⁸ were coined during the Octavianus period (40 BC), first in Lugdunum and later in other mints in northern Italy. As the necessity for local production increased for political reasons, the coins showed large differences in style and morphology; at first they were well minted and had

* Correspondence to: I. Calliari, Department of Mechanical and Management Innovation, University of Padua, Via Marzolo 9, 35141 Padua, Italy

[†] Presented at the European Conference on EDXRS, Lisbon, Portugal, 23–29 June 1996.

Contract/grant sponsor: CNR, 'Progetto Strategico Beni Culturali'.

Table 1. Compositions (wt%) of RIC99, RIC329 and CRA471 determined using EDXRS, EDS and AAS on the surface (s) and on bulk (b)

Coin	Component	EDXRS (s)	EDXRS (b)	EDS (b)	AAS	
RIC99	Fe	0.81	0.26	0.48	0.35	
	Cu	51.0	80.8	80.6	82.6	
	Zn	7.45	17.7	18.9	16.12	
	Sn	0.04	0.05	n.d.	n.d.	
	Pb	0.62	1.25	n.d.	n.d.	
		EDXRS (s)	EDS (s)	EDXRS (b)	EDS (b)	AAS
RIC329	Fe	2.76	5.47	0.51	0.79	0.52
	Cu	86.6	78.2	70.0	67.9	71.6
	Zn	9.90	11.2	26.4	30.9	27.1
	Sn	n.d.	5.10	n.d.	n.d.	n.d.
	Pb..	0.77	n.d.	0.89	n.d.	0.29
		EDXRS (s)	EDXRS (b)	EDS (b)	AAS	
CRA471	Fe	0.10	0.04	n.d.	0.02	
	Cu	80.8	74.1	74.1	71.9	
	Zn	0.32	0.24	n.d.	n.d.	
	Sn	0.66	0.36	1.92	1.44	
	Pb	17.8	24.2	23.9	26.6	

a standard weight, but progressively the quality decreased, resulting in coins of low weight and poor stylistic quality.

Twelve Julius Caesar orichalcum coins

The Julius Caesar coins (46 BC) were minted in Rome after the oriental triumphs and represented the first orichalcum coinage; this new alloy is very different from the Cu–Pb–Sn alloy and from pure Cu previously employed for asses. Julius Caesar ordered the use of this alloy and personally controlled the emission of the Rome mint and the supplies of zinc.

The Octavianus and Julius Caesar emissions were clearly in contrast to emphasize the political contrast between the two rulers.

In order to correlate composition and microstructure, in a previous study⁹ three coins representative of different alloys (Cu–Pb and Cu–Zn) with different surface conditions were analysed: a Claudian sesterce, minted in Rome (41–42 AD), RIC 329; an Augustan sesterce, minted in Rome (18 BC) in the name of T. Quinctius Crispinus Sulpicianus, RIC 99; and an as issued under Cnaeus Pompeius, minted in Spain (45–43 BC), CRA 471. The codes RIC99, RIC329, CRA471 (RIC means Roman Republican Coinage, CRA means Crawford Roman Republican Coinage), taken from the numismatic classification, are maintained here so that numis-



Figure 1. Light micrograph of RIC329 (after etching): twinned grain structure. Field: 900 × 750 μm).

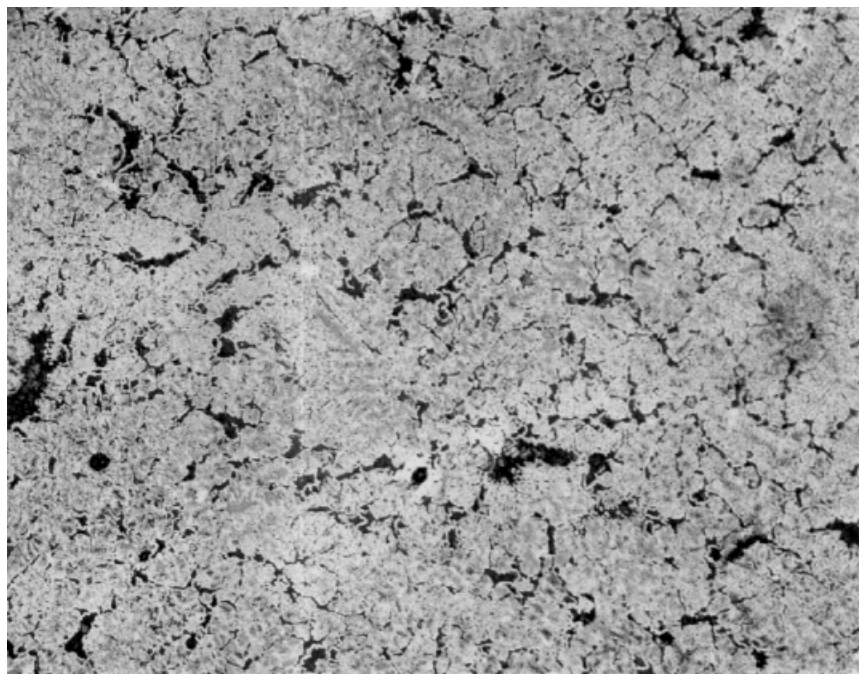


Figure 2. Light micrograph of CRA471 (after etching): Cu matrix with Pb segregated at grain boundaries. (Field: $900 \times 750 \mu\text{m}$).

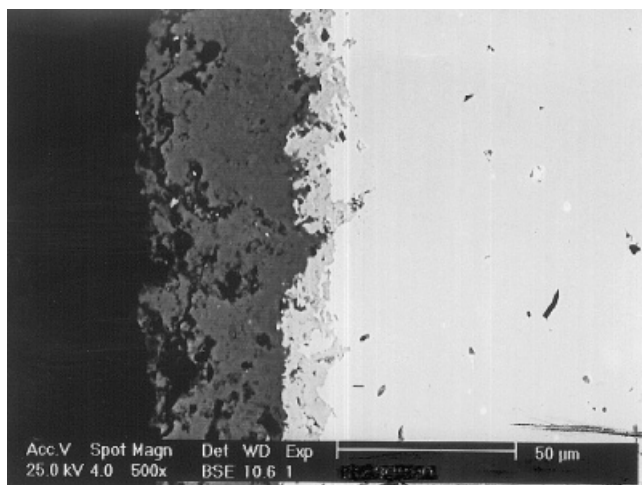


Figure 3. BSE image of a cross-section of RIC99 with a surface-modified layer of about $70 \mu\text{m}$ (dezincification process).

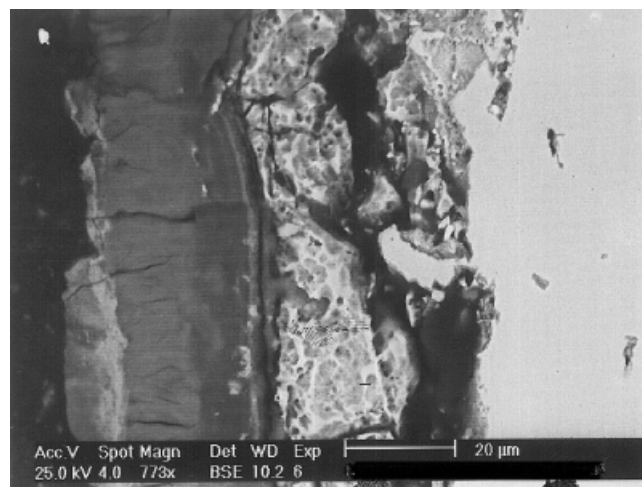


Figure 4. BSE image of a cross-section of RIC329 with a modified surface layer of about $60 \mu\text{m}$.

matic researchers will always be able to find the composition of each coin, even if it has been destroyed.

Methods

In the present study, x-ray microanalysis (EDS) and x-ray maps were also performed on the same coins to determine the element distribution and the concentration profile from the surface to the bulk. The alloy microstructures were investigated using optical microscopy on polished and etched surfaces.

Morphological examinations, microanalyses and x-ray maps were carried out on cross-sections obtained from the coins labelled RIC99, RIC329 and CRA471. Four Octavianus asses (Ot2, Ot3, Ot8, Ot38) were abraded (depth about $300 \mu\text{m}$) on the edge (area about 2 mm^2) in order to determine the bulk compositions by EDXRS and EDS.

Quantitative analysis was performed standardless with ZAF correction¹⁰ and by scanning the electron beam across a $0.2 \mu\text{m}^2$ sample area.

For AAS analysis 300 mg of bulk alloy were dissolved.

All the other coins were examined with EDXRS without any polishing treatment.

For EDS and x-ray maps the SEM was operated at 25 kV.

For EDXRS determinations the fundamental parameter method was applied. The spectrometer operated under the following conditions: Ge secondary target for Fe, Cu and Zn, 15 kV, 0.5 mA, 300 s; Gd secondary target for Pb, Au, Ag and Sn, 55 kV, 1.0 mA, 300 s.

To verify the accuracy of the method, Cu–Zn and Cu–Pb–Sn standard alloys were analysed under the same experimental conditions as for coins. The calculated concentration values agreed within 5% with the concentrations of the certified samples.

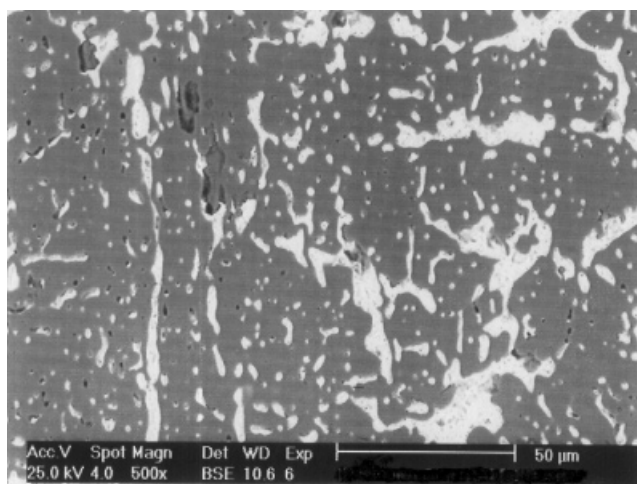


Figure 5. BSE image of a cross-section of CRA471 showing the immiscibility of Cu (dark zone) and Pb (light zone).

RESULTS AND DISCUSSION

In Table 1 the compositions (wt%) of the three coins which were cut, determined using AAS, EDS and EDXRS are reported.

In Figs 1 and 2, the microstructures of RIC329 (Cu–Zn) and of CRA471 (optical microscopy) are shown, and Figs 3–5 report the backscattered electron (BSE) images of cross-sections of RIC99, RIC329 and CRA471.

RIC329 consists of a single-phase alpha-brass¹¹ (see EDXRS analysis) with a twinned grain structure and some Pb segregation islands. Considering that the grains appear not to be strain-hardened and that the grain size is not uniform, it can be concluded that the coin was probably annealed after coining.

The BSE image (Fig. 4) shows a modified surface layer about 60 μm thick, where x-ray microanalysis and x-ray maps indicate a non-uniform composition: Si, and

Fe are localized on the external layer of about 20 μm (clay deposit); Zn is absent for 50–60 μm; Cu is quite uniformly distributed underneath the Si- and Fe-rich layer. This is due to the dezincification process,¹² which generally takes place for Cu–Zn alloys containing more than 15% of Zn. In this process most Zn is selectively removed from the alloy, leaving behind a porous and weak deposit of nobler Cu and CuO.

The bulk composition determined by AAS, EDXRS and EDS is subject to a relative error of <10%. The composition determined from the analyses on the surface (Table 1, RIC 329) and the x-ray maps confirm the qualitative data on the dezincification process.

The BSE image of RIC99 (Fig. 3) shows a modified surface layer about 70 μm and the x-ray microanalysis and x-ray-map evidenced that for 50 μm underneath the surface Zn is virtually absent; this is due to the above-explained dezincification process. The thickness and form of corrosion depend on the chemical and physical properties of the water and/or soil in which the coin was buried.

The micrograph of CRA471 (Fig. 2) and the BSE image (Fig. 5) show a Cu matrix with Pb segregated at grain boundaries. The composition data from the surface and the bulk, determined by different methods are reported in Table 1 (CRA 471). EDXRS and EDS data agree with AAS for the bulk, while the surface composition differs from the bulk about by 10% for Cu and 25% for Pb.

As can be seen, there is good agreement between the data concerning the bulk obtained by AAS, EDXRS and EDS. The lower relative content of Pb on the surface layer can be attributed to its insolubility in the Cu matrix and to its lower melting point, both of which account for its migration towards the bulk upon solidification.

In Table 2 are reported the compositions (wt%) of the surfaces of the Julius Caesar coins determined with EDXRS. Most of the Julius Caesar coins were not very damaged, so that the surface composition provides rea-

Table 2. Compositions (wt%) of 12 Julius Caesar coins (EDXRS on surface)

Component	JC1	JC2	JC3	JC4	JC5	JC6	JC7	JC8	JC9	JC10	JC11	JC12
Fe	9.48	0.35	0.27	0.6	0.86	2.0	0.5	1.52	0.16	0.59	0.4	0.19
Cu	79.1	80.4	86.1	85.6	88.7	89.1	74.5	85.4	74.5	84.5	79.5	77.8
Zn	17.7	17.7	11.9	13.4	9.51	6.75	24.5	10.6	23.6	12.5	19.1	21.2
Ag	0.10	0.10	0.07	0.09	0.10	0.12	0.07	0.12	0.08	0.02	0.08	0.10
Pb	0.68	0.68	0.44	0.71	0.61	1.82	0.48	1.98	0.43	2.40	0.92	0.61
Au	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	1.2	n.d.	n.d.	n.d.
Sn	0.76	0.76	1.15	0.08	0.13	0.19	0.05	0.36	0.02	0.02	0.05	0.05

Table 3. Compositions (wt%) of nine Octavianus coins (EDXRS on surface)

Component	Ot1	Ot1 bis	Ot4	Ot6	Ot7	Ot10	Ot34	Ot37	Ot44
Fe	0.48	0.76	1.67	0.43	0.91	0.17	0.24	0.59	0.47
Cu	74.6	78.9	54.5	67.5	70.8	72.0	65.6	80.1	72.4
Zn	0.20	0.34	0.25	0.28	0.30	0.32	0.28	0.10	0.28
Ag	0.09	0.12	0.10	0.20	0.36	0.11	0.09	0.07	0.07
Pb	17.4	10.8	35.5	29.7	23.4	21.1	24.4	11.5	18.3
Au	0.53	0.42	0.16	1.29	1.07	0.71	0.78	0.59	0.49
Sn	6.55	8.61	7.73	0.58	3.54	5.55	8.82	6.66	7.95

Table 4. Compositions (wt%) of Ot2, Ot3, Ot8 and Ot38 determined with EDXRS and EDS on the surface (s) and on bulk (b)

Coin	Component	EDXRS (s)	EDS (s)	EDXRS (b)	EDS (b)
Ot2	Fe	5.91	19.8	5.20	0.14
	Cu	59.2	26.7	67.7	82.7
	Zn	5.73	17.7	4.42	0.60
	Ag	0.16	n.d.	0.09	n.d.
	Pb	11.0	16.4	13.6	8.86
	Au	0.57	n.d.	0.68	n.d.
	Sn	17.4	19.6	8.28	7.69
Ot3	Fe	0.38	1.54	0.38	0.43
	Cu	88.6	87.6	85.9	87.2
	Zn	6.56	7.07	7.08	9.14
	Ag	0.33	n.d.	0.81	n.d.
	Pb	1.95	2.57	2.71	1.68
	Au	0.87	n.d.	0.81	n.d.
	Sn	1.28	1.25	2.30	1.58
Ot8	Fe	0.16	1.01	0.27	0.25
	Cu	77.7	54.0	74.8	81.4
	Zn	0.31	n.d.	0.35	n.d.
	Ag	0.07	n.d.	0.08	n.d.
	Pb	12.4	30.1	16.1	10.7
	Au	0.38	n.d.	0.78	n.d.
	Sn	8.94	14.9	7.61	7.59
Ot38	Fe	0.19	0.68	0.16	0.14
	Cu	88.1	92.0	87.5	90.4
	Zn	0.51	1.86	0.58	0.70
	Ag	0.17	n.d.	0.13	n.d.
	Pb	2.63	4.51	4.14	4.34
	Au	0.51	n.d.	0.55	n.d.
	Sn	5.53	0.97	3.85	4.37

sonable information on the bulk composition; the data obtained confirm the numismatic data about orichalcum coinage.^{4,13}

Table 3 summarizes the compositions (wt%) of the surfaces of nine Octavianus asses, obtained with EDXRS. The composition of these coins is close to the traditional Cu–Pb–Sn alloy employed for asses issued, for example, by Sextus Pompeius.^{8,14} A low-quality Cu-based alloy was employed, for both emissions, with high

Pb and Sn content. The octavianus coins were officially minted in Lugdunum and in several 'private' mints in Italy, which accounts for the differences in alloy composition.

In Table 4 the surface and bulk compositions of four Octavianus asses obtained with EDXRS and EDS are reported. These data suggest that the composition obtained by EDXRS on the coin surface satisfactorily reproduces, from the numismatic point of view, the composition of the whole coin.

CONCLUSIONS

The microstructure examination supported the chemical analysis data, explaining compositional changes from the surface to the bulk, in particular concerning Pb and Zn. The compositions determined by AAS show a satisfactory correlation in particular with the EDXRS data.

When examining high Pb-containing materials, a strong migration and/or segregation effect was noticed, which accounts for the large differences between the EDS results (very small investigated volume) and the 'bulk' techniques EDXRS and AAS.

The analytical determinations confirm the numismatic hypothesis that the coins issued by Julius Caesar were made of orichalcum alloy and the asses issued by Octavianus and Sextus Pompeius were of Cu–Pb–Sn alloy.

EDXRS provides a good compromise between the need for a representative analysis and the necessity to preserve the integrity of the coins and, as a first step, when the research has undertaken to examine a vast range of coins in order to determine which kind of alloy was employed. Other techniques have to be used when an accurate chemical analysis of selected samples is requested.

Acknowledgements

Financial support by the Italian National Research Council (CNR, 'Progetto Strategico Beni Culturali') is acknowledged.

REFERENCES

1. J. Bayley, *B. Mus. Occas. Pap.* No. 50, p. 7. British Museum, London (1990).
2. G. F. Carter, *Isr. Numism. J.* **6–7**, 22 (1982–83).
3. E. R. Caley, *Numism. Notes Monogr.* **151**, 73 (1963).
4. J. Riederer, *Jahrb. Numism. Geldgeschichte* **24**, 7 (1974).
5. G. F. Carter, *Ame. Numism. Soc. Mus. Notes*, **33**, 91 (1988).
6. G. F. Carter, *Archaeometry* **7**, 106 (1965).
7. L. A. Glinsman and L. C. Hayek, *Archaeometry* **35**, 46 (1983).
8. R. Martini, *Sextus Pompeius*, Glauk I. Ennerre, Milan (1995).
9. I. Calliari, M. Magrini and R. Martini, *Sci. Techn. Cult. Heritage in press*.
10. J. A. Small, in *Handbook of X-Ray Spectrometry: Methods and Techniques*, edited by R. Van Grieken and A. A. Markowicz, p. 606. Marcel Dekker, New York (1994).
11. *Metals Handbook* Vol. 9, p. 404, ASM, New York (1995).
12. D. A. Jones, *Principles and Prevention of Corrosion*. Macmillan, New York (1992).
13. U. Zwicker, in *Proceedings of the XIth International Numismatic Congress*, edited by T. Hackens and G. Moucharte, p. 73. Louvain-la-Neuve (1993).
14. M. Amandry, J. N. Barrandon and J. C. Richard, *Rev. Archeol. Narb.* **19**, 57 (1986).