INVESTIGATIONS ON ANCIENT INDIAN METALLURGY

I. A Pre-historic Bronze Bowl

By S. PARAMASIVAN

(Archæological Chemist, Government Museum, Madras)

Received December 6, 1940

Introduction

METALS are the physical sinews of modern civilisation. The knowledge of the complexity of combination of physical, mechanical and chemical properties which is demanded of a given metal for a particular use is an index to modern civilisation.

Metals and knowledge of them occupied almost the same position with the historic and pre-historic civilisations of the past. Some of the pre-historic art-ware and domestic utensils are still the wonder of the world. But to a student of chemistry, the knowledge of commercial alloys used in pre-historic times, and the basic processes and problems involved in their utilisation and their relation to accepted principles of modern physical metallurgy form a fascinating study. In reconstructing some phases of ancient civilisation and thereby helping archæological research, metal objects thus afford important clues.

These facts have been fully realised in the West* where the British Association appointed the "Sumer Committee" to investigate, among other things, on the metal objects of ancient Mesopotamia, and probably also of Egypt. No one has so far attempted to do anything like it in metal-lurgy and metallography to reconstruct the exact methods of fabrication of historic and pre-historic metal objects in India.

In India, pre-historic civilisation existed at Mohenjo-Daro and Harappa and other parts of Sindh and the Panjab between the 4th and the 3rd millenniums B.C., which had commercial and cultural intercourse with Mesopotamia. In addition there were the pre-historic civilisations existing in Adichanallur,

^{*} Sir Harold Carpenter was the first to show the way in his investigations on an Egyptian bronze spear head, vide Nature 1931, 127, 589-91; 1932, 130, 625-27.

Coimbatore and other sites in S. India, which probably had connections with the civilisations of Mohenjo-Daro and Harappa. And an investigation like the one here is bound to yield fruitful results in many directions.

The Madras Government Museum has a very large collection of prehistoric bronze objects from Adichanallur¹ and Coimbatore² probably dating from the 2nd or the 1st century B.C. In order to reconstruct the ancient metallurgy and the exact method of workmanship of these objects, a bronze bowl from Coimbatore collections was taken up for investigation.

The bronze bowl which is typical of Coimbatore and Adichanallur collections of similar objects has an internal diameter $12 \cdot 1$ c.m. and a depth of $4 \cdot 5$ c.m. Portions of it are broken, probably through cracking.

Experimental

The results of chemical analysis of the bronze bowl are as follows:—

		Chemical Analysis (Per cent.)
Copper	• •	75.25
Tin		23.58
Arsenic	• •	tr.
Lead		0.21
Iron		0.37
Nickel	• •	0.28

The remaining 0.31% represents oxygen in the combined state occurring as cuprite.

The bowl had been covered with a green patina which was removed by means of a wire brush and dilute hydrochloric acid. Under the patina was a layer of cuprite covering the metal. This also was removed. A part of the bowl was then polished and etched with ammonium persulphate and examined under a metallurgical microscope.

At 100 magnifications, it was observed that the metal had recrystallised and that the resulting crystals exhibited the following characteristics:—

- (i) There is a eutectoid complex of two phases forming islands, bounded by well-defined band of bluish-white constituent.³
- (ii) The eutectoid complex is not composed of equi-axed grains, but are elongated.

From the constitutional diagram⁴ of copper-tin alloys, it will be seen that the structure of these alloys containg more than 12% of tin depend upon the heat treatment they have received. Above 500° C. they are composed of α and β phases,* a structure possessing great strength and permitting forging at temperatures within the α - β range. This structure can only be preserved by cooling repidly from above 530° C. Slowly cooled alloys of the same composition do not contain the β -constituent, but break up into the eutectoid complex of the α and δ phases⁵* forming islands bounded by well-defined band of the hard bluish-white δ constituent. This substance, sometimes regarded as a definite compound Cu_4Sn is a source of brittleness and alloys containing it are weaker than those of the same composition in which the α - β structure has been preserved by quenching.

The next point to be considered is the elongation of the grains. They are obviously the deformed equivalents of the previously equi-axed crystals through external stresses. The manner in which such deformation of a crystal can occur is readily understood when it is realised that in any crystal which is built up by the arrangement of the atoms (or molecules) on some regular space lattice, there must be certain planes upon which displacement under external stress can take place without any disarrangement of the atomic distribution. From a geometrical point of view, this could occur on every plane parallel to two of the main lines of space-lattice. If the displacement occurred in steps which are equal to or are exact multiples of the atomic spacing in the direction of displacement, then such displacement would leave the entire crystal arrangement unchanged, although the external shape of the crystal would be altered. By numerous displacements of this kind, however, a very large degree of deformation of the crystal as a whole could be brought about.

The deformation and elongation of the grains could have taken place here only in the course of forging the material to shape, and not through mere casting.

That the object was not cast but forged to shape and cold worked is also proved thus. The Vickers Diamond Hardness Number (which is almost equal to the Brinell Hardness Numbers) varied from 190 at the bottom of the bowl to 263 over the rim, while the hardness of the middle portion was 221. Thus different portions of the bowl have been subjected to different degrees of mechanical treatment.

^{*} Vide Pl. LXXXIII—Guillet and Portevin, Metallography and Macrography (G. Bell and Sons, Ltd., London, 1922).

2. Rea, A.

Since the crystals do not appear to be well developed and have eutectic structure, they have not been properly annealed. But such severely cold worked condition is characterised by a high yield point and great hardness, but the elastic limit is usually low. They should never be put to work in this overstrained condition. They should be subjected to low temperature heat treatment, which removes the internal strain. The heat treatment raises the elastic limit and restores the elastic properties, at the same time increasing slightly the yield point and the hardness. But the microscope fails to distinguish between overstrained objects and the same objects aged or restored by low temperature annealing. Thus it is possible that some such low-temperature annealing should have taken place, though it is impossible to recognise it microscopically. But the cracking and disappearance of a portion of the bowl may be the result of "season cracking" caused by internal stress due to cold work and subsequent insufficient annealing.

The alloy has retained much of the hardness imparted by cold work, but it is difficult to say whether it has lost a portion of its original hardness in the course of centuries.

REFERENCES

1. Breeks, J. W.

An Account of the Primitive Tribes and Monuments of the Nilgiris (London, W. H. Allen & Co., 1873), Appendix C, pp. 126-37.

.. Catalogue of the Pre-historic Antiquities from Adichanallur and Perumbair (Government Press, Madras, 1915), pp. 4, 8-11.

3. Desch .. Metallography (London, Longmans, Green & Co., 1922), p. 391.

4. Guillet and Portevin .. Loc. cit., p. 228.

5. Desch .. *Loc. cit.*, p. 61.

6. Greaves and Wrighton .. Practical Microscopical Metallography (London, Chapman & Hall, 1933), p. 110.

II. Ancient Indian Bronze Coins of the 2nd and 11th Centuries A.D.

General

Ancient Indian kings minted bronze and copper coins and the extant specimens of them housed in museums of the world date from about the 4th century B.C. to almost the modern times. But no attempt has so far been made to study their exact fabrication. Recently, investigations were conducted by the author on an Andhra coin of about the 2nd century A.D. and a Chola coin of the 9th century A.D. They were about 7 mm. in diameter and 3 mm. in thickness. Of these two, the Chola coin was more copper coloured showing that it was richer in that metal.

Experimental

The chemical analyses of the two coins are as follows:—-

	-	Chemical Analyses (Per cent.)	
		Andhra Coin 2nd C.A.D.	Chola Coin 9th C.A.D.
Copper	 	76.69	96.40
Tin	 • •	18·14	2.13
Antimony		1.66	tr.
Lead	 	0.31	1 · 47
Iron	 	3 · 20	tr.
Arsenic		tr	tr.

The Andhra coin contains a large percentage of tin and antimony and hence must be harder than the Chola coin, which is richer in copper. The presence of antimony in the Andhra coin is significant. It gives a better cast. Lead and iron are probably impurities, but the latter might have been purposely added to the Andhra coin.

The two coins were covered with a thin green crust of corrosion products and underneath them was a layer of cuprite. The two layers were removed by means of wire brush and dilute hydrochloric acid. They were polished and etched with ammonium persulphate and their structure examined under a metallurgical microscope.

At 100 magnification, both coins showed that,

- (i) the metal had completely crystallised;*
- (ii) the crystals were not, in any manner, deformed, elongated or twinned.
 - (iii) the grains had fine structure;
- (iv) there were no columnar grains¹ (in other words the crystal growth had no preferred orientation).

This structure showed that the coins were cast and not forged to shape and that there was no cold working. The absence of cold working is further proved thus. Under cold working, different portions of the surface will have different degrees of hardness. But the hardness of the two coins was uniform throughout their surface, being 231 and 91 (in Vickers Diamond Hardness Scale)† respectively for the Andhra and the Chola coins. Thus the former was harder than the latter and better able to withstand the wear in circulation.

The fine grained structure is the result of a high rate of solidification. A high temperature warms the mould before solidification commences, and the rate of solidification is low. Thus castings poured at too high a temperature generally show a coarse mechanical structure and are weak mechanically.

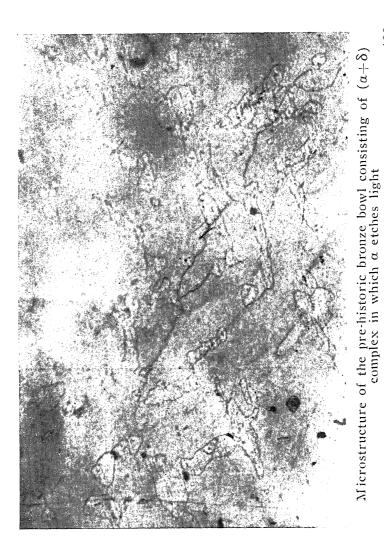
It is evident from the fine structure that the casting was not poured at too high a temperature and that the rate of solidification was high. The ancient mint masters should have used either a metal mould or a sand mould with metal chills, or a wet clay mould. It is evident from the proportion of copper and tin that the Andhra coin should have been heated to a temperature of not less than 900° C. and the Chola coin not below 1100° C. before molten liquid was poured into the cast.

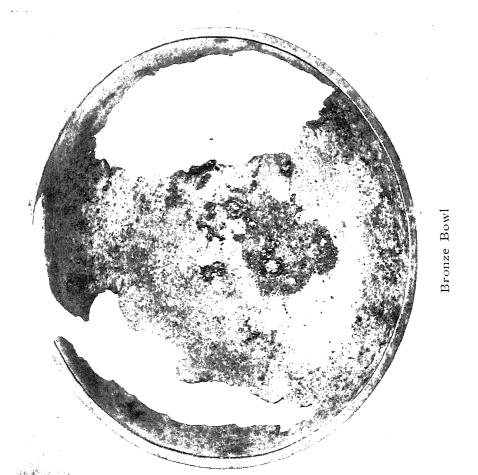
We do not know why the Cholas preferred a softer alloy for the coinage and why the art of coinage which was superior under the Andhras deteriorated.

In conclusion, the author desires to express his thanks to Mr. R. Lean, Chief Mechanical Engineer and to Mr. G. C. Mills, Chemist and Metallurgist of the M. & S.M. Railway, for permitting him to do the work in their Chemical

^{*} The Chola coin had a 'cored' structure, in which the corrosion product is seen to coincide with the dark 'cores'.

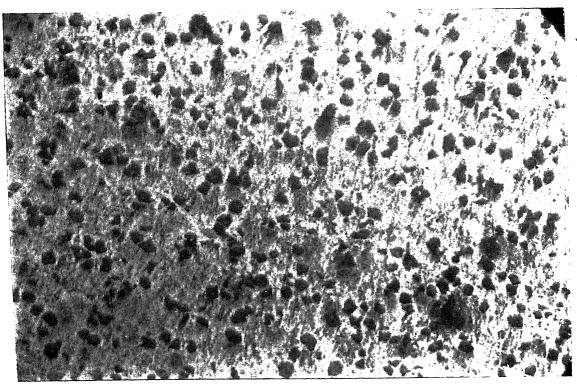
[†] The Vickers Diamond Harndess figures almost closely agrees with Brinell Hardness figures.







Microstructure of the Chola coin showing 'cores' corresponding to corrosion products and consisting of α (light) solid solutions × 100



Microstructure of the Andhra coin consisting of α (dark) and $(\alpha+\delta)$ complex in which δ etches light

190

and Metallurgical Laboratories. His thanks are also due to Mr. K. P. S. Nair, Ag. Chemist and Metallurgist of the M. & S.M. Railway, with whom the author has discussed some of these problems and to Dr. F. H. Gravely for his keen interest in the subject.

REFERENCES

1. Doan .. Principles of Physical Metallurgy (New York, McGraw Hill Book Coy., 1935), p. 31.

2. Glazebrook, R. .. Dictionary of Applied Physics (London, Macmillan & Co., 1923), p. 359.