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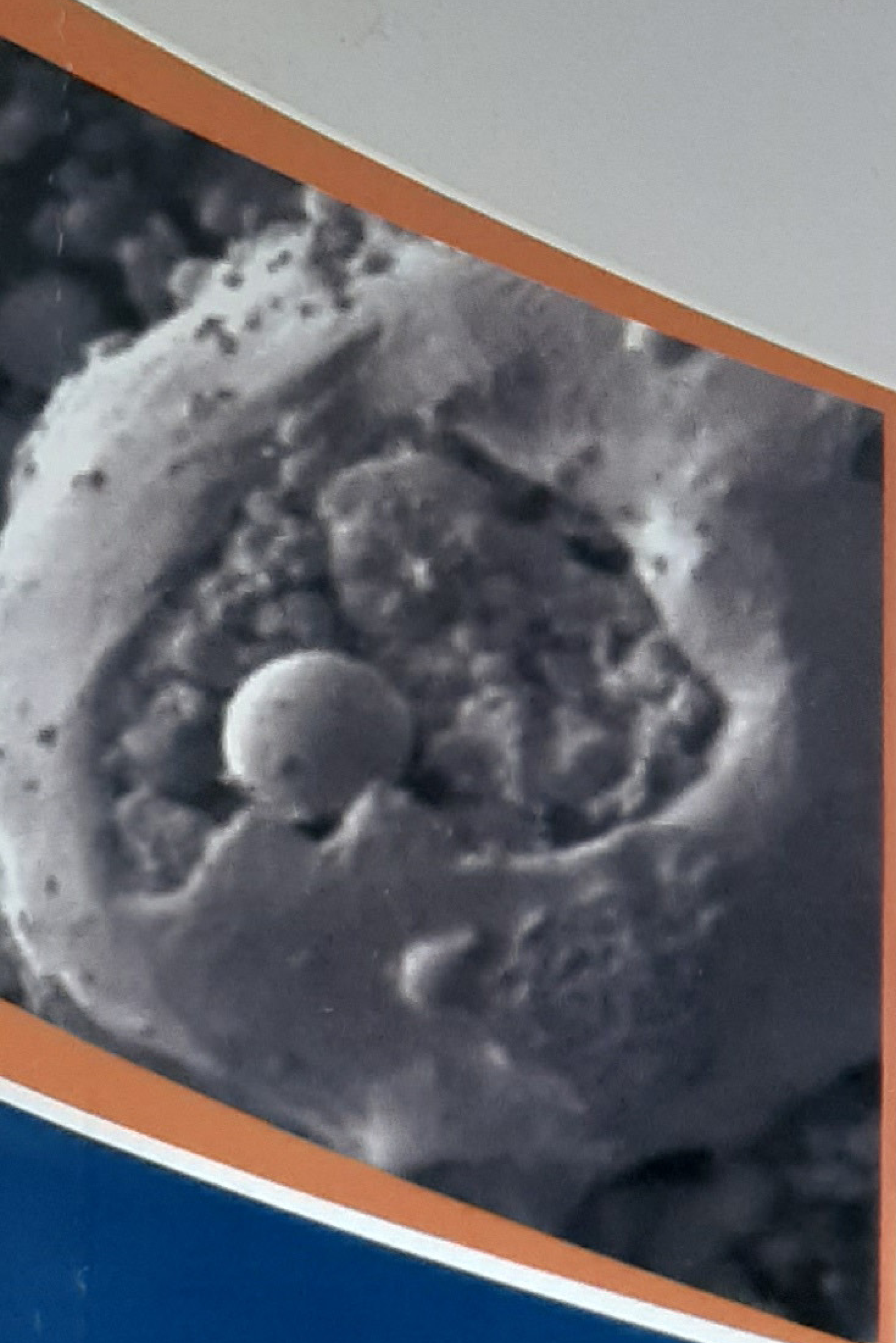

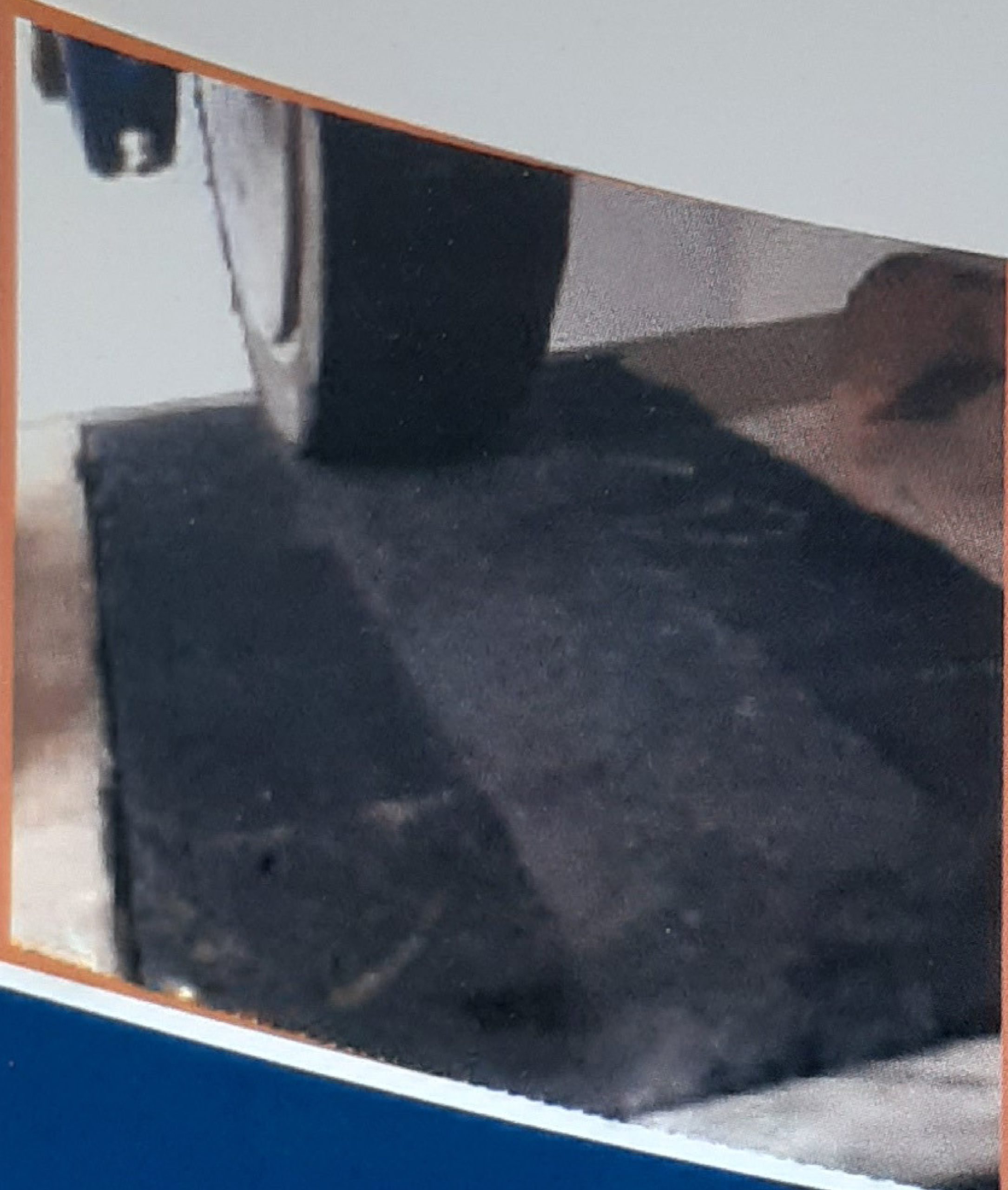

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Key Metals and Alloys from Indian and South Indian Antiquity: An Archaeometallurgical Overview

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1 INTRODUCTION

Archaeometallurgy is increasingly becoming a widely inclusive discipline of archaeology which attempt to trace the history of ancient metal production, distribution and usage in antiquity and the related socio-cultural and economic ramifications (Srinivasan 1996). Metals have played a crucial role ever since post-Neolithic societies being used progressively through the Copper Ages, Bronze Ages and Iron Ages across different regions. The early metals to be exploited were those which were found in the native state, followed by those which could be smelted or reduced easily from ores, while those which were more difficult to smelt were discovered last. The commonly used metals in antiquity include gold, silver, copper, iron, tin, lead, zinc and mercury (Srinivasan and Ranganathan 1997). Many modern developments in metallurgy draw from in ancient practices that pre-date the Industrial Revolution. The earliest usage of copper seems be around the 8th millennium BCE in Turkey or Anatolia (Rehren and Pernicka 2008). The use of non-ferrous metals is seen in the river valley cultures of Mesopotamia, Egypt; and also the coeval Harappan civilisation of the Indian subcontinent (c. 2500 BCE) (Possehl and Rissman 1992, Kenoyer 1998).

The trajectory of some of the metals used in antiquity is traced in this paper with some archaeometallurgical insights, field studies and technical investigations by the author, particularly from southern India. Various techniques of scientific examination find increasing use in archaeometallurgical and archaeometric study, including microscopy, spectro-chemical and elemental analysis, x-ray fluorescence and others. Investigations of archaeometallurgical debris such as slags, crucible and furnace remnants can be made using SEM-EDAX, EPMA-WDS and so on. Lead isotope ratio analysis can be a useful technique for identifying the source of lead alloyed in an artefact since the lead isotope ratios remain unchanged through smelting processes (Srinivasan 1999a), which has been attempted by the author on some artefacts as reported here.

2 GOLD IN ANTIQUITY

The noble metal gold is found in nature in the native state. Gold has been used to make jewellery not only due to its golden lustre but also due to its great ductility which facilitated forging it into sheet metal and a range of shapes. The most spectacular gold artefact is the Egyptian artefact of the enigmatic mask of the young Pharaoh Tutenkhamen (c. 1300 BCE) made by hammering sheet metal. Early gold and silver ornaments from the Indian subcontinent are found from Indus Valley sites such as Mohenjodaro (c.3000 BCE). Gold and copper usage is reported from Neolithic Merhgarh in Baluchistan, Pakistan (c. 6000 BCE). Diadems and belts of gold are reported from Mohenjodaro. Harappan silver artefacts are reported from Kunal, in north-western India (Agrawal 2000: 6). The lighter colour of some gold artefacts from Mohenjodaro suggests the use of naturally occurring gold with silver impurities. Skilled practices of goldworking are noted in the Harappan period such as the use of gold micro-beads (Figure 1).

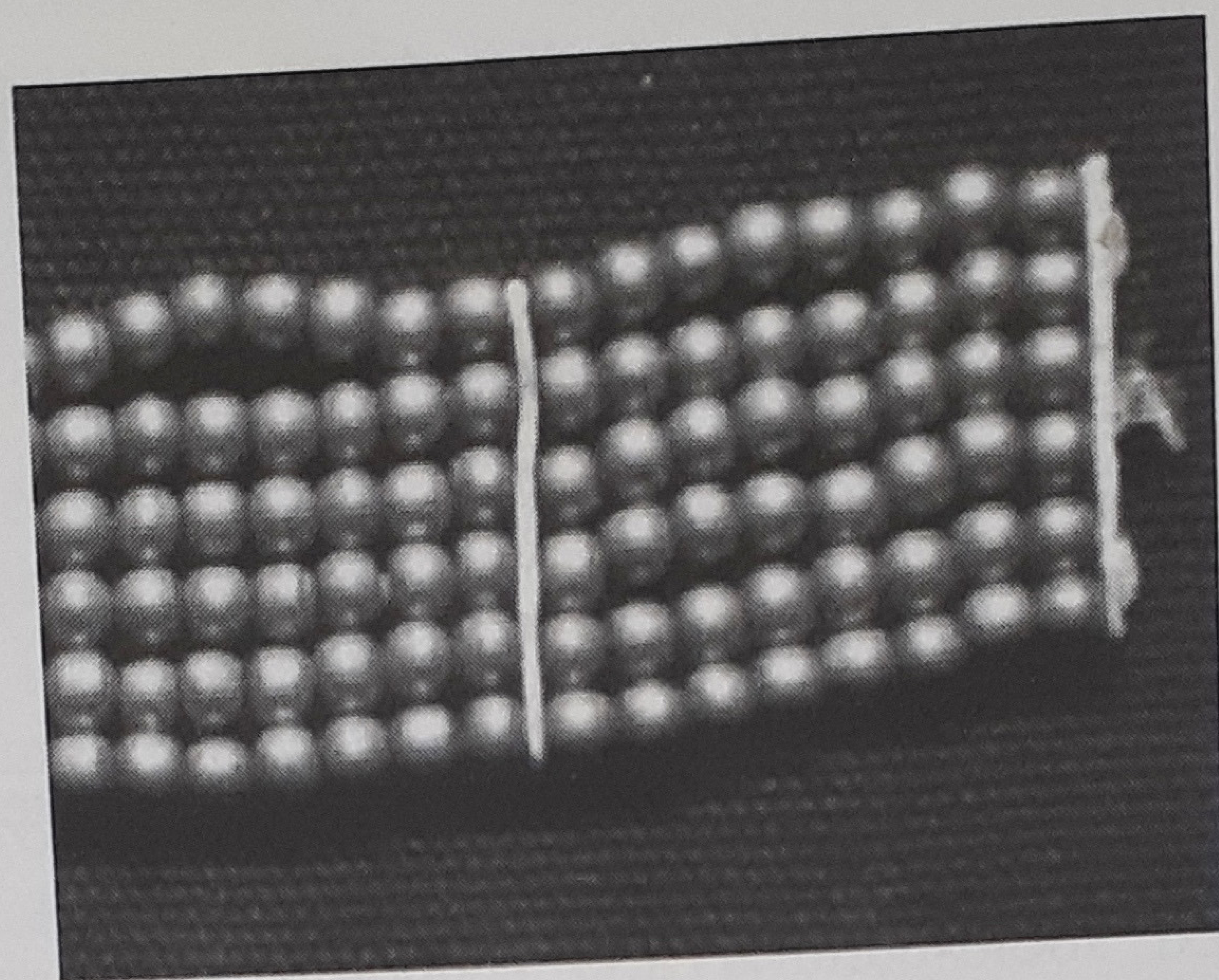

Figure 1: Micro-beads from Harappan site, National Museum, Delhi (photo credit: Sharada Srinivasan)

Gold mining seems to have had a long history in parts of southern India such as in Karnataka. It has been speculated that gold from the Karnataka region collected from the surface by Neolithic cultures of the mid third millennium BC might even have supplied the Indus regions (Allchin and Allchin 1982: 337). More recent studies have also pointed to other probable nearer minerals sourced for the Indus period (Law 2011). The author had made preliminary field surveys in north Karnataka in the Hutti-Maski region in 1991 where she noted extensive old workings for gold. Most outcrops had open cast mines with old mining galleries with large mullacker fragments scattered about indicating ore crushing activity in antiquity. Old timber from a 200 metres deep mine shaft was carbon dated the mid 4th century BC (Radhakrishna and Curtis 1991: 23-4), ranking amongt the deepest known old gold mines. The Jalagarus were a traditional community in the Dambal region who undertook alluvial washing and panning for gold (Foote 1874: 140).

The Nilgiri hills and Wynad bordering, Tamil Nadu, Kerala and Karnataka host some sparse hard rock and alluvial gold deposits. Roman Pliny's account (1st century) of gold from the country of Naris might well refer to the land of the Nairs, ie the region of the alluvial gold tracts of the Nilambur valley below the Wyand hills (Radhakrishna and Curtis 1991: 23). While they are currently uneconomical, it is interesting they have been illegally mined/panned by local Kurumba tribes as observed by the author. In 1990, the author and Digvijay Mallah had identified some old gold workings in Gudalur (Figure 2), and observed children from the local Kurumba community engaged in hard rock mining for gold and panning for gold from the streams for alluvial gold using large wooden pans (Figure 3), whereby the heavier particles of gold would segregate into the pan while the lighter sand grains would wash away (Srinivasan 2016a). The Kurumba tribe was traditionally believed to have had magical powers apart from knowledge of mining and metallurgy according to Thurston (1909). There are also accounts that Hoysalas used gold from the region.

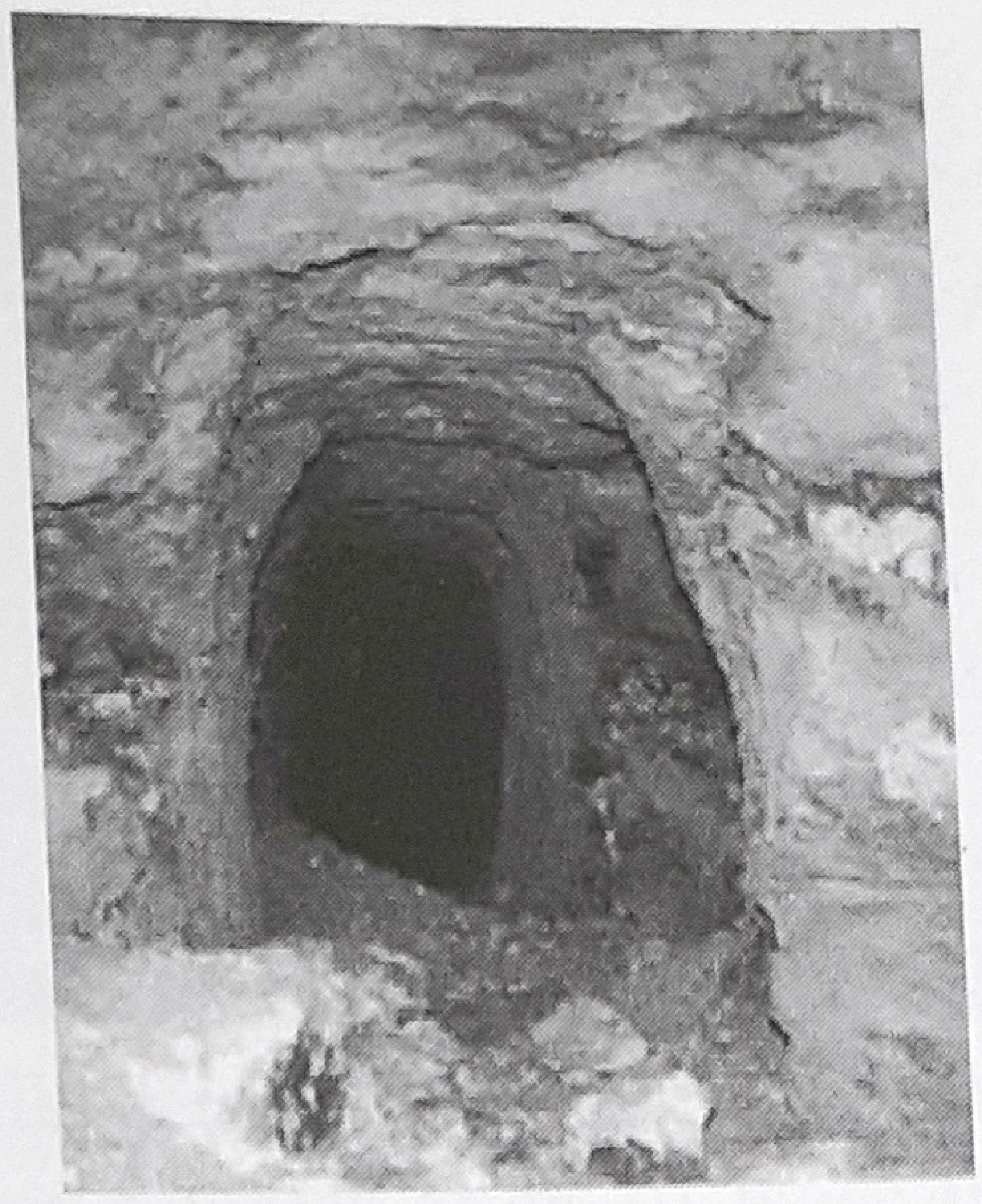

Figure 2: Old gold working at Gudalur, Nilgiri district, Tamil Nadu, 1991 (photo credit: Digvijay Mallah)

Figure 3: Panning of gold by Kurumba children at Gudalur, Nilgiris district (1991) (photo credit: Digvijay Mallah)

The rich finds of gold jewellery from the Nilgiri cairns, now housed in the British Museum, London may date from the early or mid 1st millennium BC to AD by some commentators (Knox 1985). The gold granulation technique seen in some of the ear-rings, whereby tiny spheres of gold were formed, may relate to Hellenistic influences, although the use of gold micro-beads was also noted at Harappan sites such as Lothal. Other early Tamil examples of gold jewellery include a ear-ring from Souttoukeny of the 2nd century BCE from Tamil Nadu depicting a prickly fruit, now in the Musee Guimet in Paris (Postel 1989: 130). These bring to mind the rich poetry of the classical Tamil Sangam era (c. 3rd century BCE to CE) which evoke local fruits and flowers such as kurinji.

3 SILVER

Silver was extracted in the Old World using the method of cupellation, by the smelting and refining of silver rich lead ores. The old mines and working in Dariba and Agucha in the region of Rajasthan indicate production of silver from argentiferous lead in antiquity. These mines were found to be comparable in extent to the mines of Rio Tinto in Spain used in the Classical and Hellenistic World (Craddock et al. 2017). Silver anklets were also found from the Harappan site of Mandi. Use of silver is seen in punch-marked Mauryan coins from the 4th century BCE onwards. From the Satavahana period, (1st-2nd century CE), lead isotope finger-printing suggested the Agnigundala mine in Andhra Pradesh as one source of silver for coins, while Sardinia seemed likely to be another source, indicative of maritime trade (Srinivasan 1999b, 2016b). The largest cast silver urns are seen in the Jaipur palace and museum which would made of alloyed metal since pure silver is too soft for such large castings.

4 CAST COPPER-BRONZE TOOLS AND ICONS

The use of copper-bronze tools is seen from Harappan times, ranging from utilitarian artefacts such as chisels, nails, hooks and axes to cast miniature figurines. Several examples of low tin bronze with less than about 10% tin are found, whereby tin would have been added to harden the softer copper metal and to improve its castability. The fine miniature bronze of the Mohenjodaro dancing girl is about 10 cm in height. Cast in the round, it was very likely made of the lost wax/resin casting process where a model would have been made of wax/resin, invested with clay to make the mould and the wax melted out and metal poured into the hollow. Mortimer Wheeler in a TV programme of 1973 described the image as, 'a girl, perfectly confident, there is nothing like her in the world' (Possehl 2002). The casting of copper alloy icons came widely in vogue from the early historic period onwards. Excellent examples of Gupta statuary are known such as the life-size Sultangunj Buddha now in the Birmingham Museum, which was found to be of predominantly of copper and standing at 2.28 m. Spectacular bronzes were also cast under the Cholas in southern India. The celebrated Nataraja bronze of the Chola period (Figure 4), was hailed as 'poetry but nonetheless science' by Ananda Coomaraswamy in 1912. Bronzes continue to be cast by the lost wax process even in the present day at Swamimalai, in Tanjavur district (Figure 5). In this process an image was made of wax and invested with clay to form the mould. The mould was then heated to melt and get rid of the wax and then metal poured in which solidified to give the final metal icon. The hollow casting process used a clay core and was used more in the north of India for casting. In this the final icon was made of a thin layer of metal with the clay core retained inside. Hollow cast icons can appear damaged due to the thinner layer of metal being prone to damage.

Figure 4: Nataraja, Kankoduvanithavam, Government Museum, Chennai (photo credit: Benoy Behl)

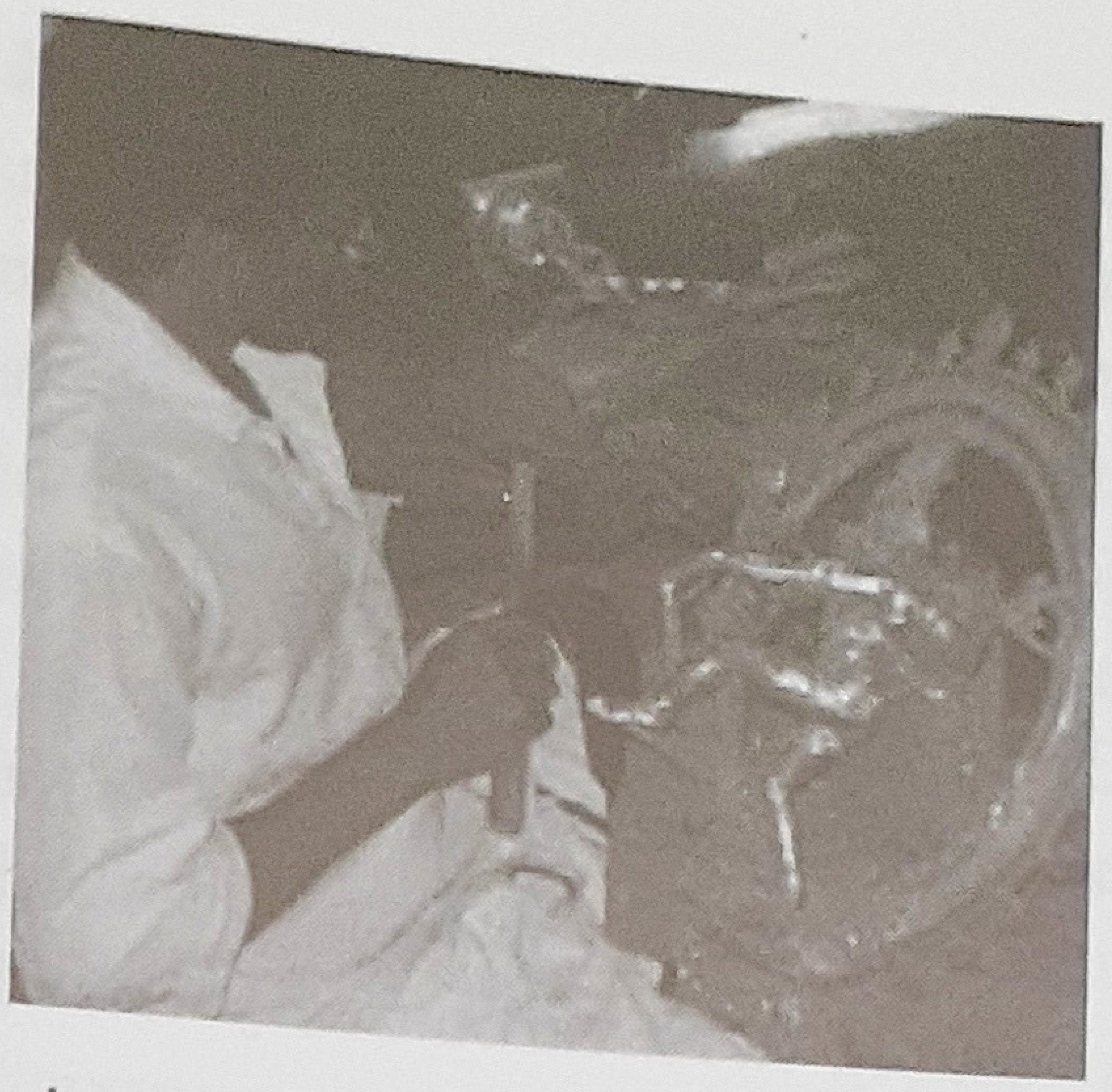

Figure 5: Icon maker at Swamimalai, Thanjavur district, Tamil Nadu (photo credit: Sharada Srinivasan)

Harle (1992: 302) memorably commented that the early Chola bronzes represent the finest representations of godhood, unsurpassed in any place or age. These bronzes were made by the lost wax process or investment casting process. The image was cast by first making a model in wax, and then invested with moulding material to form a mould and thereafter the mould is heated to expel the wax and the molten metal is poured into the hollow to generate the metal icon. The Sanskrit phrase *'madhuchchehistavidhanam'* refers to the lost wax process and is described in the artistic treatise of the Manasara of about the 4th-5th century (Reeves 1962: 29-31, Srinivasan 2016c). That the lost wax process is also invoked in a poem by the Tamil women poet-saint Andal (*c.* 800) who compares dark rain clouds to the mould holding liquid wax, entreating them to rain on the Lord Visnu (Srinivasan 2016c).

Investigations by the author as previously reported in Srinivasan (1996, 2016) indicate that a majority of the South Indian medieval bronzes were of leaded tin bronze. About 80% of 130 south Indian images from the early historic to late medieval period were leaded bronzes with tin contents not exceeding 15% and keeping within the limit of solid solubility of tin in copper. Beyond this limit as-cast bronzes become increasingly brittle due to the increasing presence of delta phase. The Chola bronzes (10th -12th century) in general had an average composition of about 6% tin and 6% lead (Srinivasan 2015). Of the total number of images only 15% were leaded brass images with more brass being used in the post-Chola period. Archaeometallurgical investigations by the author on slag specimens recovered near the Ingaldhal copper mines in Karnataka confirmed that they were from copper smelting, likely of the Satavahana period from associated finds of early historic russet coated pottery (Srinivasan 2016d).

5 USE OF BINARY HIGH-TIN BRONZES

As-cast binary copper-tin alloys with over 15% were not widely used in the ancient world as they are embrittled due to the presence of the delta phase component. Previously, the Indian subcontinent had not been regarded as a significant region in the exploitation of tin and bronze. However, metallurgical investigations by the author on artefacts from megalithic contexts and early historic contexts, continuing into medieval to modern south India demonstrated longstanding familiarity with the exploitation of the intermetallic properties of binary high-tin bronzes, as seen in the manufacture of vessels, coins and musical instruments of wrought and quenched beta bronze with 22-5% tin, and the manufacture of mirrors of delta bronze with about 33% tin (Srinivasan 1994, Srinivasan and Glover 1995, Srinivasan 2016e), which are also the last surviving crafts of their kind in the world.

Astonishingly, highly sophisticated and thin-rimmed bronze vessels have been uncovered from the megalithic cairns and burials of the Nilgiris (Figure 6) and Adichanallur, Tamil Nadu (*c.* 1000-500 BC), while metallurgical investigations on some of these by the author confirmed that they were of hot forged and quenched binary unleaded high-tin beta (23% tin) bronze (Srinivasan 1994, Srinivasan and Glover 1995, Srinivasan 2010). Such an alloy of copper and tin with around 23% tin can be forged greatly at high temperatures due to the presence of a high temperature plastic beta phase which when quenched gives additionally properties of strength and lustre to the alloy. These extraordinarily thin-rimmed vessels were fabricated by extensively hammering out such an alloy between 586-798°C when a plastic beta intermetallic compound (Cu_5Sn) of equilibrium composition of 22.9% tin forms. This was followed by quenching which resulted in the retention of needle-like beta phase (as seen in the microstructure of a vessel from Adichanallur in Figure 7) and prevents the formation of brittle delta phase and also gives a golden polish. Low-tin bronzes have limited workability in comparison. Indian influences were also discerned in examples of high-tin bronze vessels found in Thailand in southeast Asia (Bennett and Glover 1992)

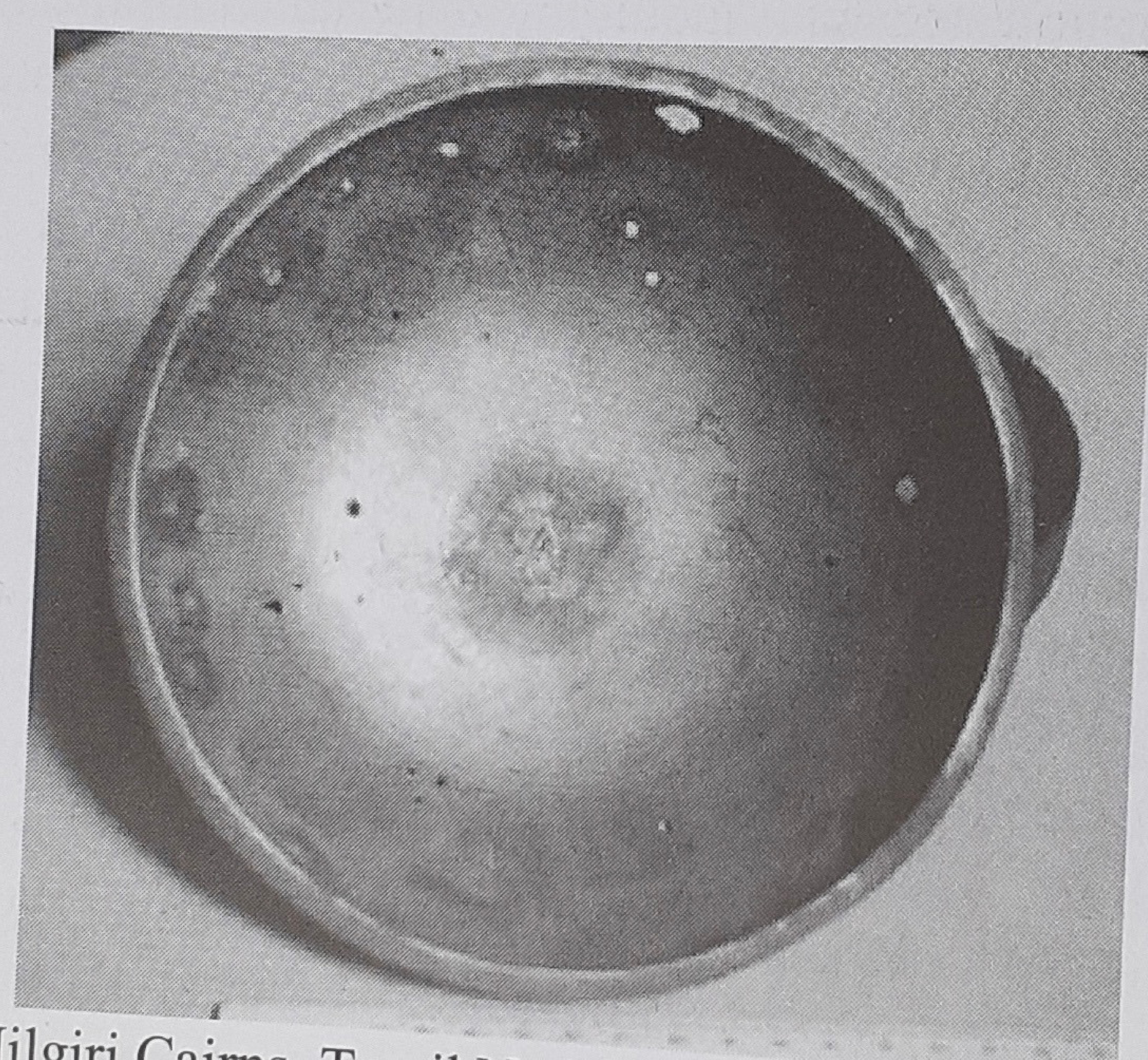

Figure 6: Vessel from Nilgiri Cairns, Tamil Nadu, Government Museum, Chennai (early to mid 1st mill BCE) (photo credit: Sharada Srinivasan)

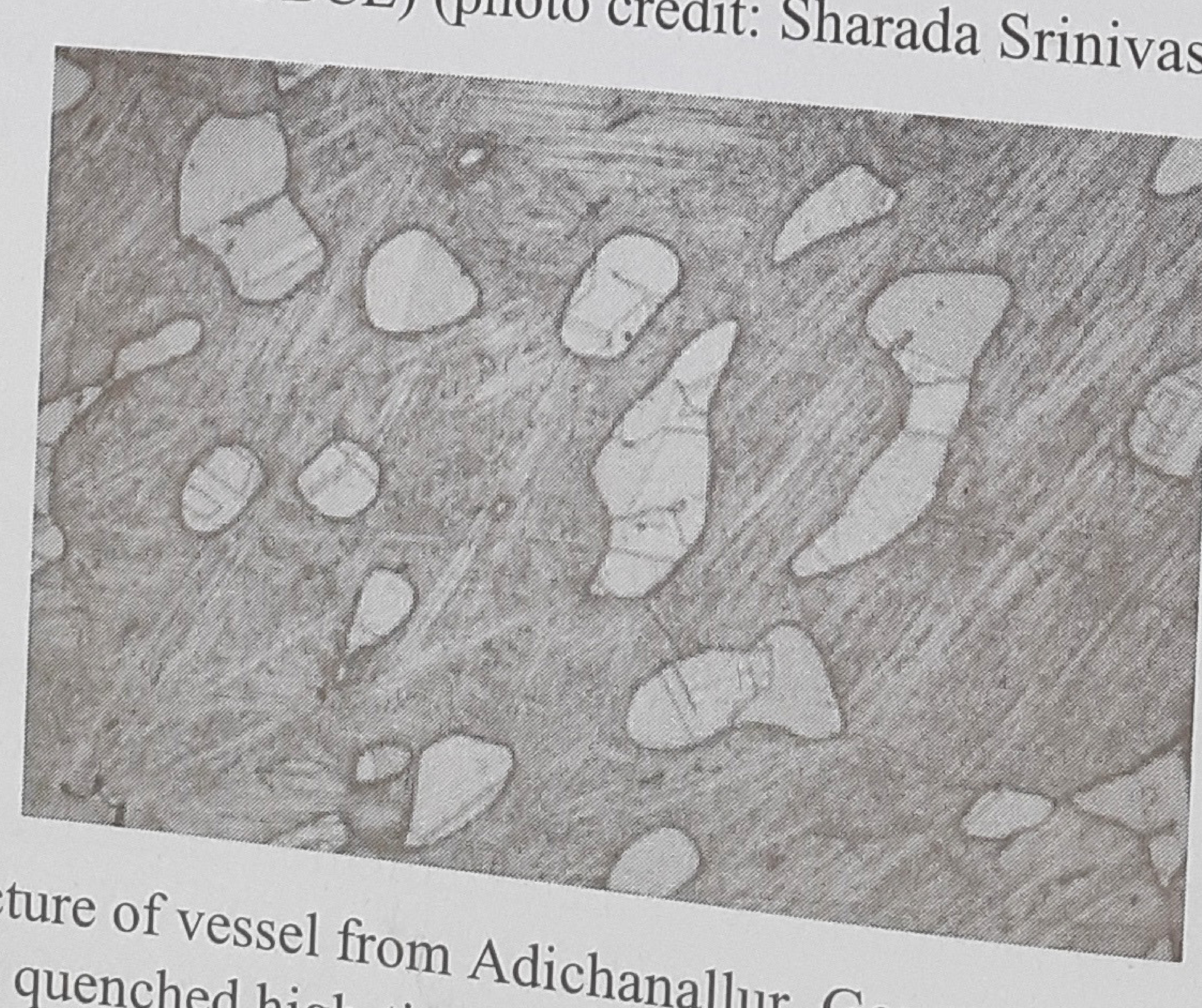

Figure 7: Micro-structure of vessel from Adichanallur, Government Museum, Chennai, hot forged/wrought and quenched high-tin beta bronze structure of 23.9 % tin bronze, 400X (photo credit: Sharada Srinivasan)

Such high-tin bronze vessels have continued to be used among the local communities of the Nilgiris such as the Todas. Such high-tin beta bronze vessels also show high corrosion resistance due to the retention of the beta intermetallic compound phase as also seen in the Nilgiri vessel in Figure 6. The making of such high-tin bronze vessels by similar processes survived in many places till recently, such as in Kerala and in Nacharkoil in Tamil Nadu. The author observed large vessels being made in parts of Kerala especially in Palghat district in the 1990s of 23% beta bronze, 25 cm in diameter and 1 mm rim thickness,being wrought and hot forged from ingots of 15 cm diameter and 1.5 cm thick followed by quenching. However this tradition has virtually died out today.

Although tin is scarce in India compared to other regions such as southeast Asia, it is possible that some minor local tin deposits were accessible in antiquity. Eastern India has tin deposits in the Hazaribagh region (Chakrabarti 1979, 1985-6), where Mallet observed the pre-industrial smelting of tin by local tribals in furnaces resembling shaft furnaces for iron smelting. Investigations by the author on slags from the ancient mining region of Kalyadi within Hassan district of Karnataka indicate that these are bronze smelting slags with up to 7% tin from co-smelting copper and tin ores due to the presence of metallic iron, rather than casting slags from alloying copper and tin (Srinivasan 1997), which might suggest the exploitation of minor local sources of tin.

6 EARLY PRODUCTION OF METALLIC ZINC

The earliest firm evidence for pre-industrial extraction of metallic zinc seems to comes from India. Zinc is one of the most difficult of metals to isolate since zinc metal oxidizes and volatalises readily around the same temperature of about 1000°C as is needed to smelt zinc ore. Hence it forms as a vapour in the furnace which would immediately get re-oxidised. Therefore finds of metallic zinc are not very common from early antiquity.

There is remarkable evidence for the semi-industrial extraction of metallic zinc by the 12th century CE from the Zawar area of Rajasthan (Craddock et. al. 1998). Zinc was smelted by downward distillation of the zinc vapour formed after the reduction of zinc ore. The *Rasaratnakara,* a Sanskrit text ascribed to the great Indian scientist Nagarjuna, of the early Christian era describes the process of downward distillation or *tiryakpatana* (ibid.). Using retorts with condensers and specially designed perforated furnaces, the zinc vapour could be drastically cooled down to about 500° to collect at the bottom of the furnace to get a melt that could solidify into zinc metal. Remnants of perforated furnaces with zinc smelting retorts have been found from Zawar, whereby more than 30 retorts seen to have been packed in each furnace (Craddock *et al.* 1998, Craddock *et al.* 2017). The remains (Figure 8) suggest that production of Zawar was almost on semi-industrial scale in an era preceding the Industrial Revolution and continued on a large scale during the Moghul era until the 17th century. A sample with 34% zinc wasexcavated from the Buddhist site of Taxila (ca 4th c. BC) or Takshashila, now in Pakistan (Marshall 1951) which may have been made by alloying metallic zinc.

A metallic zinc ingot with a reported Deccan Brahmi inscription was studied by the author using lead isotope ratio finger-printing and was found to fit a 5th c. AD attribution from the Andhra region (Srinivasan 1998, 2016a). The shape of the ingot/coin was also interesting akin to a solidified globular droplet with a flat bottom as it could have been collected at the bottom of the furnace by downward distillation. In Europe, commercial zinc smelting operations were established by William Champion in Bristol in Britain in the 1740's using downward distillation suggesting its inspiration from the Zawar process (Craddock et. al. 1989). Thus Indian metallurgists can justifiably be seen to have invented the process of zinc smelting.

The remarkable artistic innovation of Bidri ware, inspired by Persian inlaying traditions, developed under the late medieval Muslim Sultanate rulers of theBidar region of Karnakata. The use of metallic zinc was made to make highly elegant metalware, of a patinated high-zinc alloy with 2-10% copper inlayed in silver (La Niece 2015), which was used to make hukka bases, ewers and other artefacts.

Figure 8: Zinc smelting retorts from Zawar, Rajasthan (Photo credit; Paul Craddock)

7 IRON, STEEL AND WOOTZ STEEL

The Indian subcontinent has a vibrant iron and steel in antiquity. The celebrated Iron Pillar (Figure 9) is renowned as the 'rustless wonder' for its relative corrosion resistance. It is about 7.375 m high and 41.6 m in diameter and is the earliest surviving massive iron forging. The Sanskrit inscription on the pillar of the late 4th century to early 5th century CE is attributed to the Gupta king Chandragupta II, alluding to the erection of a *dhwaja* or pillar by Chandra, as a devotee of the Hindu deity Vishnu, on the hill of Vishnupadagiri. A sample studied by Sir Robert Hadfieldwas found to be iron of high purity with about 0.1% phosphorus and 0.04% carbon (Hadfield 1912). The formation of a protective passive film on the surface and an amorphous oxyhydroxide layer next to the metal-rust surface of the phosphoric iron may have aided the corrosion resistance (Balasubramiam 2008). Ultrasound investigations suggest that the pillar was built up by forge-welding cakes of wrought iron in a perpendicular and radial fashion (Raj *et al.* 2005).

Figure 9: Delhi Iron Pillar, 4th century, upper portion (Photo credit: Digvijay Mallah)

India has also been famed for the legendary Indian wootz steel, or *ukku*in south Indian languages, a high-grade high-carbon steel, especially produced in southern India according to several Europeantravelers' accounts from the 16th-17th centuries onwards (Bronson 1986).As indicated in accounts of 17th century traveler Tavernier, tens of thousands of shipments of wootz steel from sent from the kingdom of Golconda (in modern Telangana) to Persia and West Asia to make the fabled Damascus blades. The Damascus steel blade, believed to have been forged of Indian wootz steel of a high carbon content of 1.5-2%, was reputed for its cutting edge (Smith 1982, Srinivasan 1994, Srinivasan and Ranganathan 2014). The attempts to characterize wootz by scientists of the caliber of Michael Faraday spurred many developments in 19th century metallurgy and contributing to the Industrial Revolution. Vast amounts of archaeometallurgical debris related to the production of wootz steel are still found in the region of northern Telangana. Wootz steel production sites were also uncovered in the region of Mel-siruvalur and Tiruvannamalai in Tamil Nadu by the author (Srinivasan 2016f, Srinivasan 2007).

As far as the archaeological evidence is concerned, three specimens of ferrous blades had high carbon contents of 1.2-1.7% C of the composition of wootz steel from the Bhir mound of Taxila (*c.* 3rd century BCE (Marshall 1951: 534). Greek alchemist Zosimos in the 3rd c, AD mentioned that the Indians made steel for sword by melting soft iron in cruciblesrecalling to the wootz process (Craddock 1995: 279). Kadebakele is an Iron Age site near the World Heritage site of Hampi in Karnataka. A small iron ring (acc. no. 900, 22E-28 N, Level 7) radiocarbon dated to 800–440 BCE yielded a through pearlitic structure of at least 08% carbon steel, suggesting it could have been cast steel produced from crucible processes (Srinivasan *et al.* 2009).

Excavations at an iron age megalithic site at Kodumanal, Tamil Nadu (3rd *c.* BCE), near Karur, the Chera capital of the Sangam era (3rd c. BCE-3rd c. AD) revealed furnaces with vitrified crucibles (Figure 10) and iron slag (Rajan 1990). A vitrified crucible fragment from Kodumanal showed ferrous metal processing remains (Srinivasan and Griffiths 1997, Srinivasan 2007). Figure 11 is an elemental distribution map of a crucible fragment from Kodumanal, examined using EPMA-WDS by the author which shows the presence of ferrous remnants. The bardic poems of the SangamTamil poetess (3rd c. BCE-3rd c. AD) evoke warring chieftains and their spears (*ekku*). *Ukku* may derive from the Sangam Tamil word *uruku*, meaning boiling over. The Sanskrit Arthasasthra (3rd c. BCE) refers to *vaikruntaka* which might possible allude to steel. Pliny's 'Natural History' mentions the import into the Roman world of iron from the 'Seres', which might be identified with the ancient Southern Indian kingdom of the Cheras. Roman accounts of the 'Periplus of the Erythrean Sea' refer to the flourishing port of Muziris or Muciri on the Malabar coast. Interestingly, an iron nail from Pattanam, identified with the Muziris revealed a microstructure typically associated with ultra-high carbon wootz steel containing about 1.5% C (Srinivasan 2007).

Figure 10: Crucible and tuyere fragments, Kodumanal, 3rd century BCE (Photo credit: Digvijay Mallah)

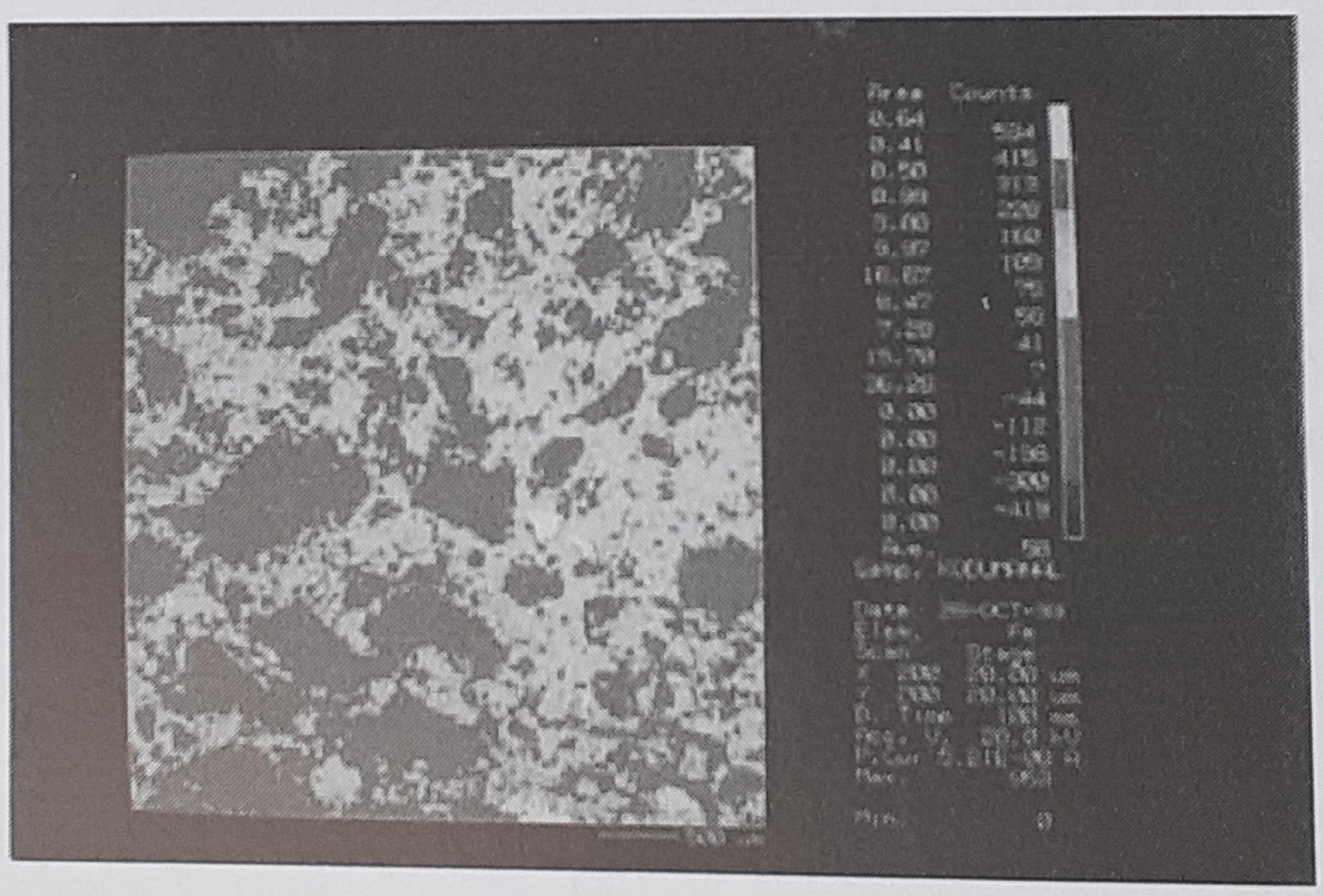

Figure 11: Elemental distribution map by EPMA-WDS on cross-section of crucible from Kodumanal, 3rd century BCE, Tamil Nadu (Photo credit: Sharada Srinivasan)

8 ARCHAEOMETALLURGY AND CONSERVATION SCIENCE: CHALLENGES AND SCOPE

In the Indian context there are several challenges in developing the fledgeling area of archaeometallurgy. Sadly, many of the surviving crafts traditions linked to ageold practices are rapidly declining with livelihoods increasingly marginalised. Several of the archaeometallurgical production sites and old mining areas are also disrupted by infrastructure development, agriculture and so on, so that the records of these activities are rapidly being effaced. Concerted action is needed to retrieve what is left of the remnants of a rich pre-industrial legacy. The area of conservation research is also one that needs more impetus in terms of scientific research. Even though the Delhi Iron Pillar, has been hailed as the rustless wonder, a closer look (Figure 9) suggests that in recent times rust has perhaps indeed been forming perhaps exacerbated by industrial pollution and other factors. Thus, there is a need for working concertedly for the scientific documentation and preservation of artefacts and materials heritage.

9 ACKNOWLEDGEMENTS

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