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Khao Sam Kaeo – an archaeometallurgical crossroads for trans-asiatic technological traditions

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1. Introduction

Whilst cultural transmissions across the Bay of Bengal from the mid 1st millennium CE are undisputed 'facts' of Southeast Asia's past, the prehistory of these interactions is an ascendant field of enquiry (e.g. Bellina, 2007; Bellina and Glover, 2004; Bellina and Silapanth, 2008; Glover, 1990a). Some scholars (e.g. De Casparis, 1983) had posited that the trans-Asiatic encounters responsible for the flores-cence of Southeast Asia's historical polities must have had anteced-ents for which we lack textual evidence, but it was Ian Glover (1983, 1990a, 1990b) who first attempted an account of prehistoric regional relations with the subcontinent; largely derived from his excavations in west-central Thailand at Ban Don Ta Phet. Finds of what were thought to be characteristically South Asian agate, carnelian, and glass ornaments (Glover, 1996), as well as high-tin bronze bowls (Bellina, 1999; Bennett and Glover, 1992; Rajpitak and Seeley, 1979) supported the argument that the 'indianisation' phenomenon could

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ABSTRACT

Recent archaeological investigations at Khao Sam Kaeo, on the Upper Thai-Malay Peninsula, have furnished evidence for a mid/late 1st millennium BCE cultural exchange network stretching from the Indian subcontinent to Taiwan. Typological, compositional and technological study of Khao Sam Kaeo's copperbase artefacts has identified three distinct copper-alloy metallurgical traditions, with reasonable analogies in South Asian, Vietnamese, and Western Han material culture. Furthermore, analyses of technical ceramic and slag suggest that Khao Sam Kaeo metalworkers may have been using a cassiterite cementation process to produce high-tin bronze ingots for export or onsite casting/forging. Not only would this industry constitute the earliest evidence for the exploitation of Peninsula tin resources, but we also offer a speculative argument for the source of Khao Sam Kaeo's copper-base production technology.

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have been underway by the mid 1st millennium BCE (e.g. Bellina, 2007, 2001; Bellina and Glover, 2004; Bellina and Silapanth, 2008; Rispoli, 2006, 2005; Srinivasan, 1998; Glover, 1990b).

It was formerly assumed that prehistoric material culture with South Asian attributes were simply sporadic imports from South Asia, and that Southeast Asian social groups unquestioningly adopted their 'Indian' materiality (sensu Martinón-Torres and Rehren, 2009; Miller, 2005; Taylor, 2008), but subsequent research has enriched our appreciation of the social significance and multilateralism of so-called 'indianisation' processes (e.g. Bellina, 2007; Bellina and Glover, 2004). Technological analysis of carnelian and agate ornaments from across South and Southeast Asia (founded upon an ethnoarchaeological study of a hard-stone bead industry with reasonable historical continuity in Khambhat, Gujarat, India: see Roux, 2000; Roux et al., 1995) resulted in the identification of artefacts from mid/late 1st millennium BCE contexts in Thailand, Vietnam, and as far as the Tabon Caves in the Philippines, all bearing stigmata of highly skilled South Asian production techniques (Bellina, 2001, 2003, 2007). Critically, the realisation that ornaments with distinctly Southeast Asian typologies had been formed by complex 'Indian' techniques suggested that trans-Asiatic flows of cultural information had been multidirectional, and that South Asian, or South Asian-trained, artisans

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adapted their production to fulfil Southeast Asian demand – within the latter's aesthetic/ideological repertoire (*ibid*.). The traditional unilateralist perspective of 'indianisation' was further upset by the recognition of production debris at Khao Sam Kaeo (hereafter 'KSK') on the Upper Thai-Malay Peninsula. The ethnoarchaeological data (Roux, 2000; Roux et al., 1995) strongly suggest that such highly skilled hard-stone knapping and polishing skills would require lengthy apprenticeships to master. This leaves two scenarios: either Southeast Asians were trained on the subcontinent for extended periods or, as Bellina (2001) proposed, small numbers of South Asian artisans may have settled at KSK, arguably to better respond to Southeast Asian desire for high quality ornaments conveying or emphasising elevated social status.

Not only did this study extend the sphere of prehistoric subcontinental cultural influence from the Thai-Malay Peninsula far into the South China Sea, it both overlapped and supported previous proposals (e.g. Bellwood, 1984–1985, 1985, 1991; Bellwood et al., 1995; Solheim, 1961a, b, 1966, 1984, 1988, 2000, 2006) for 'Austronesian groups' circulation and/or exchange networks (Fig. 1). Recent compositional studies suggesting that Fengtian (Taiwan) nephrite roughouts and finished artefacts were also circulating from east to west, as far as peninsular Thailand at least, further highlights the reciprocity of material and social exchanges around the South China Sea (Bellina and Silapanth, 2008; Hung et al., 2007); and the latest data (expanded upon below) would now incorporate Western Han Chinese interactions over the same geographical range at the turn of the 1st millennium BCE/CE (Bellina and Silapanth, 2008; Pryce et al., 2008, pp. 304–305 [and references therein]). Thus, the combined data indicate a segmented but interlocking trans-Asiatic sphere of prehistoric maritime interaction spanning in excess of 5000 km.

Located at the narrowest point of the northern Thai-Malay Peninsula, the Kra Isthmus, the settlement and industrial centre of KSK constituted a physical and cultural crossroads between the Andaman Sea and the Gulf of Thailand from the 4th century BCE to the 1st century CE (Fig. 2a). An overland route via broadly contemporary west coast sites like Phu Khao Thong and Ban Kluai Nok with South Asian-related material culture could potentially save over a thousand kilometres of maritime passage via the Straits of Melaka (cf. Bellina and Silapanth, cf. 2008, pp. 262-263; Jacq-Hergoualc'h, cf. 2002,p. 83). The site covers c. 50 hectares over four hills on the eastern bank of the Tha Thapao River, c. 8 km upstream from the present shoreline of the Gulf of Siam (Fig. 2b). The heavy erosion of a dynamic monsoon environment and the ravages of modern looting ensure that KSK's stratigraphy is a frequent source of headaches, but survey and excavation since 2002 have discerned coherent evidence for earthworks, occupation, and industrial activities; with archaeobotanical and palaeoenvironmental studies ongoing. The distribution of hard-stone, glass, and metal working debris at KSK appears to indicate the presence of craft production quartiers, with metallurgical ceramics and slags concentrated on and around Hill 3 (Fig. 2b). These quartiers could relate to ethnic as well as artisanal identities (Bellina, 2008, p. 265); a hypothesis supported by a recent GIS analysis demonstrating artefact patterning to be statistically significant (Malakie, 2008). Investigation of KSK's multiple strands of technological and anthropogenic landscape have contributed to the ongoing rebuttal of 'indianisation' as a foreign imposition by emphasising the selective adoption and adaptation of exogenous material culture and materialities by Southeast Asian social groups actively involved in local, regional, and inter-regional economic and/or political relations (Bellina, 2008).

Where does metallurgy fit within the developing KSK narrative? Concentrating on KSK's copper-alloy consumption evidence, typological and macro-technological study have identified reasonable analogies in South Asia, Vietnam and Han China; seemingly indicative of the site's metallurgical cosmopolitanism (Pryce et al., 2008, pp. 300–306), and complementary to the long available Ban Don Ta Phet evidence (e.g. Rajpitak and Seeley, 1979). The bulk of the present paper relates to the supplementation of these preliminary KSK data with compositional and microstructural analyses, and the correlation of these results with the archaeometallurgical literature



Fig. 1. Relief map of the Bay of Bengal and South China Sea theatre of prehistoric maritime exchange, with major sites mentioned in the text – 'Khambhat' is the location of Roux et al.'s (1995) ethnoarchaeological hard-stone bead study and 'Chang'an' (now Xi'an) was the capital of the Western Han empire; these locales are shown to show the potential extent of trans-Asiatic interactions. Courtesy of ESRI, elevation data derived from SRTM.



Fig. 2. (a) Relief map of the Upper Thai-Malay Peninsula and central Thailand, with major sites mentioned in the text. Courtesy of ESRI, elevation data derived from SRTM. (b) 3D relief map of Khao Sam Kaeo, with defensive and hydraulic earthworks marked. Courtesy of Julie Malakie, elevation data produced by Vincent Bernard (both of Khao Sam Kaeo Archaeological Mission).

available from South and East Asia, and other areas of Southeast Asia. We argue that the KSK data constitute a robust, though partially contingent, classificatory triangulation, demonstrating the presence of three distinct copper-alloy metallurgical traditions onsite, and thus reinforcing the Peninsula's role as a cultural crossroads. But what of the production evidence? Mirroring the stone ornament industries, were the metallurgical products also aimed at feeding trans-Asiatic exchange networks?

Of course, for the Thai-Malay Peninsula, a long-term interest has been the history of the region's formerly vast tin reserves (e.g. Bellwood, 2007, p. 277; Bronson, 1977, 1992; Jacq-Hergoualc'h, 2002, pp. 43, 45, 66, 195; Pryce et al., 2008, p. 306). Unfortunately, the destructive nature of 19th/20th century mining and smelting techniques, the sluicing of ores and the meticulous crushing and resmelting of slags, including ancient ones, dictates that the Peninsula's metallurgical past will remain hazy. Nevertheless, we present tentative evidence suggesting that KSK metalworkers were producing high-tin bronze ingots for export and/or onsite casting/ forging. We also contend that this copper-base technology has its closest analogies in South Asia, with potential ramifications for the ethnic and political structure of the settlement.

The present study had two main objectives: (1) to identify and document intersections between KSK metal artefacts' stylistic, compositional, and manufacturing characteristics, and (2) to To investigate what kind of metallurgical production activities were being carried out at KSK. To achieve the first objective we studied metallic artefacts with three different stylistic influences: Southeast, South, and East Asian; and for the second objective we examined samples of slag and technical ceramic.

2. Methodology

A stratigraphic sampling strategy of KSK's metallurgical assemblage (metal, slag, and technical ceramic) provided material for analytical study, which was carried out at the University College London, Institute of Archaeology's Wolfson Archaeological Science Laboratories. The bulk chemical composition of the technical ceramics was analysed by polarising energy dispersive X-ray florescence ([P]ED-XRF) using a Spectro X-Lab Pro-2000 (Tq-0261a algorithm). The results are qualitative due to the scanning of unprepared surfaces but permitted the sub-sampling of technical ceramics with high metal contents, which were then mounted in resin and polished to 0.25 μ m.

Optical microscopy under both plane polarised light (PPL) and cross polarised light (XPL) was used to identify areas of interest for further analyses by scanning electron microscopy with energy dispersive spectrometry (SEM-EDS), which were performed with a Philips XL30. The SEM-EDS system used an accelerating voltage of 20 kV, a working distance of 10 mm, a spot size of 5.3, and a process time 5, corresponding to a deadtime of c. 30%. Data was processed by INCA Oxford spectrometer software, outputting data as elements for metal samples, and adding oxygen by stoichiometry in ceramics and slag. Ceramic chemical compositions are quantified from 8–10 analyses of an approximate area of $600 \times 400 \,\mu\text{m}$ incorporating the smallest inclusions and trying to avoid bigger ones. All data have been normalised to 100% but analytical totals are provided.

As the detection limit of the SEM-EDS unit was c. 0.5 wt%, wavelength dispersive electron probe micro analysis (EPMA) was carried out to measure minor and trace elements in metallic

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| m 11 | |
|-------------|--|
| Table | |

Accuracy test using the standard Bronze IPT 10A, certified reference material no. 0686 used to monitor the realiability of EPMA analyses.

| | 0 | Zn | Ag | As | Ni | S | Sb | Cu | Bi | Sn | Fe | Pb | Со | Total |
|--------------|----------|-------|------|--------|-------|------|-------|-------|------|-------|-------|------|------|--------|
| | 0.56 | 4.98 | 0.00 | 0.00 | 0.34 | 0.08 | 0.10 | 83.42 | 0.00 | 5.13 | 0.23 | 5.11 | 0.00 | 99.95 |
| | 0.51 | 4.97 | 0.01 | 0.00 | 0.33 | 0.11 | 0.09 | 84.33 | 0.00 | 4.59 | 0.22 | 4.74 | 0.00 | 99.91 |
| | 0.51 | 4.96 | 0.01 | 0.00 | 0.32 | 0.08 | 0.10 | 84.51 | 0.00 | 4.66 | 0.22 | 4.49 | 0.00 | 99.86 |
| | 0.59 | 4.96 | 0.00 | 0.00 | 0.34 | 0.07 | 0.11 | 83.61 | 0.06 | 4.97 | 0.23 | 4.89 | 0.00 | 99.80 |
| | 0.54 | 4.97 | 0.01 | 0.00 | 0.33 | 0.07 | 0.08 | 84.07 | 0.03 | 4.53 | 0.23 | 4.03 | 0.00 | 98.88 |
| | 0.45 | 5.01 | 0.02 | 0.00 | 0.34 | 0.06 | 0.09 | 84.35 | 0.02 | 4.51 | 0.23 | 4.95 | 0.00 | 100.01 |
| Average | 0.53 | 4.97 | 0.01 | 0.00 | 0.33 | 0.08 | 0.09 | 84.05 | 0.02 | 4.73 | 0.23 | 4.70 | 0.00 | 99.74 |
| Standard | | 4.71 | | 0.02 | 0.33 | | 0.11 | 85.13 | | 4.58 | 0.21 | 4.72 | | |
| Absolute di | fference | -0.26 | | 0.02 | -0.00 | | 0.02 | 1.08 | | -0.15 | -0.02 | 0.02 | | |
| Relative dif | ference | -5.59 | | 100.00 | -0.91 | | 16.96 | 1.27 | | -3.30 | -7.66 | 0.36 | | |

samples. A JEOL JXA-8100 electron probe microanalyser was used, with an accelerating voltage of 20 kV and a $\times1000$ magnification corresponding to an area of 151 $\times88~\mu m$. Chemical compositions given are the average of 10 area analyses per sample. The standard Bronze IPT 10A, Certified Reference Material No 0683 from MBH[®]

Analytical Ltd., was used to monitor the reliability of the analyses (Table 1). Metallographic examinations were carried out on metal artefacts after the chemical analyses, using an alcoholic ferric chloride etchant solution and an etching time of 3 seconds, following Scott (1991, p. 72).



Fig. 3. Metal Assemblage analysed.

3. Metal artefacts

All 36 copper-base artefacts recovered at KSK were ornamental rather than utilitarian and had distinct stylistic influences (Pryce et al., 2008), from which we selected 10 trying to have a representative sample of typological variation (Fig. 3):

- MA73, is a knobbed vessel with concentric circles around a central cone. These kinds of vessels have been documented in metal, ceramic, and stone from the 3rd/2nd centuries BCE on the Indian Subcontinent (Bellina and Glover, 2004; Sing and Chattopadhyay, 2003) and have parallels with those found at Ban Don Ta Phet (Bennett and Glover, 1992; Rajpitak, 1983), at Khao Kwark in Ratchaburi Province, as well as in the Dong Son region of northern Vietnam (Janse, 1962).
- MA72 is a decorated vessel whose cast geometrical motifs echo those found on North Vietnamese Dong Son artefacts (Pryce et al., 2008). Similar decorated bowls have been recovered at Ban Don Ta Phet (Bennett and Glover, 1992; Rajpitak, 1983; Rajpitak and Seeley, 1979). From now on, we may call MA72 "Dong Son related vessel" to denote its stylistic influence, though not necessarily its provenance its provenance.
- MA77 appears to be the handle of a Dong Son drum. Supposed to have a Vietnamese or South Chinese, origin those drums have been widely discovered across insular and mainland Southeast Asia and Southern China (e.g. Bellwood, 2007; Bennett, 2008; Bernet Kempers, 1988; Calò, 2009; Cooler, 1996; Imamura, 1993; Pirazzoli-t'Serstevens, 1979; Sørensen, 1976, 1979, 1988, 1990, 1997; Srisuchat, 1993). Three drums were discovered at KSK in 1970 and two are now exhibited at Chumphon National Museum.
- MA68, a fragment of a Western Han, mirror provides the East Asian connection. Typically dated 206 BCE–9 CE, other Western Han mirrors have been found in central and southern Vietnam, but this is one of the two first reported from peninsular Thailand (Pryce et al., 2008). The typical alloy for casting these mirrors is the ternary Cu–Sn–Pb alloy with around 22–23 wt% Sn (e.g. Guiver et al., 1996–1997; Mei, 2000; Wang, 2002).
- MA35 is a thin fragment of either a vessel or, due to its morphology and composition (a ternary Cu–Sn–Pb alloy), or a bell similar to those exhibited in the Sirindhorn Anthropology Center in Bangkok.

- MA36 is a fragmentary bracelet decorated with five deep parallel grooves. Under naked eye, some polishing marks or what could be engraving or drawing lines were observed.

Three other indeterminate objects were analysed, MA4, MA 60 and MA66, which were thin fragments with incised decoration based on lines and small circles. Finally, we also selected MA78, a possible copper-base ingot that fits snugly within the 'nippled' technical ceramic vessels (see below Fig. 11).

We analysed both working techniques and chemical composition to assess the homogeneity of the assemblage, potential correlations between alloys and types or influences, and possible different provenances on the basis of their trace elements.

The first attribute of the chemical dataset that draws attention is the relatively high-tin content, which varies between 7.5 and 24.9 wt%, typically staying above 10 wt% (Table 2). Lead contents vary widely from undetected to 12 wt%. Possibly as a result of the heavily alloyed nature of the artefacts, we cannot discriminate chemical groups on the basis of their impurities, as their relative proportions may be too distorted through dilution and mixing of signatures of different metals. Although cluster analyses of the dataset separated two groups, the similarity between them was higher than 80%, the differences deriving from alloying constituents more than from impurities. Only one artefact (MA 35), containing 2 wt% of antimony, clearly stands out, whereas all the other samples show virtually no impurities at levels above 0.5 wt% (Table 2). However, two groups might be differentiated on the basis of As, Ni and Ag traces: one group with silver impurities (MA 78, 68, 72 and 36) and another one with As and Ni impurities, which always appear together (Ma 60, 73 and 78). Nevertheless, correlations between those groups and styles or working techniques cannot be made, so we should wait for the lead isotope analyses analyses to discuss any possible provenance. Two groups were defined on the basis of working techniques and correlated with the alloys chosen to produce them (Table 2): objects with lower tin contents were typically left as cast, whereas high-tin bronzes were invariably hot worked and quenched (with the exception of the ingot). These and other aspects are detailed below.

3.1. Standard tin bronzes

There is only one unleaded 'standard' bronze (\sim 15 wt% Sn), MA72 (the decorated vessel with possible Vietnamese influence).

| Table 2 | | |
|-------------------|-----------|-------------|
| EPMA Analyses and | l working | techniques. |

| MA | Zn | Ag | As | Ni | S | Sb | Cu | Bi | Sn | Fe | Pb | Со | Total | HW | Q | А | С |
|-----|------|------|------|------|------|-----|------|------|------|------|-------|------|-------|----|---|---|---|
| 35 | 0.03 | 0.10 | 0.67 | 0.24 | 0.08 | 2.0 | 80.2 | 0.05 | 7.5 | 0.13 | 8.93 | 0.02 | 98.2 | | | Х | Х |
| 68 | 0.04 | 0.11 | 0.26 | 0.12 | 0.04 | 0.2 | 68.9 | 0.04 | 24.9 | 0.20 | 5.03 | 0.04 | 99.1 | | | | Х |
| 72 | 0.07 | 0.19 | 0.02 | tr | 0.31 | nd | 84.4 | 0.14 | 14.8 | 0.03 | tr | tr | 99.8 | | | | Х |
| 72b | 0.07 | 0.18 | nd | tr | 0.67 | nd | 84.2 | 0.13 | 14.7 | 0.03 | tr | tr | 98.9 | | | | Х |
| 77 | 0.02 | 0.01 | 0.05 | 0.02 | 0.41 | nd | 76.6 | 0.05 | 10.6 | 0.14 | 11.99 | nd | 96.8 | | | | Х |
| 77b | 0.04 | 0.01 | 0.04 | 0.01 | 0.71 | nd | 80.8 | 0.04 | 10.2 | 0.13 | 7.97 | tr | 90.6 | | | | Х |
| 78 | 0.05 | 0.11 | nd | 0.01 | 0.32 | nd | 77.2 | 0.03 | 22.2 | 0.02 | 0.02 | tr | 97.5 | | | | Х |
| 4 | 0.06 | 0.01 | nd | 0.02 | 0.11 | nd | 75.9 | 0.01 | 23.6 | 0.18 | 0.01 | tr | 100.0 | Х | Х | | Х |
| 66 | 0.06 | tr | 0.09 | 0.07 | 0.13 | nd | 75.3 | 0.03 | 23.5 | 0.65 | 0.02 | 0.01 | 99.0 | Х | Х | | Х |
| 73 | 0.06 | 0.02 | 0.44 | 0.24 | 0.12 | nd | 74.5 | 0.02 | 24.0 | 0.42 | tr | 0.10 | 97.4 | Х | Х | | Х |
| 73b | 0.06 | 0.02 | 0.34 | 0.23 | 0.08 | nd | 76.4 | tr | 22.3 | 0.39 | 0.02 | 0.11 | 99.3 | Х | Х | | Х |
| 36 | 0.07 | 0.21 | nd | 0.03 | 0.16 | nd | 75.3 | 0.02 | 24.0 | 0.04 | 0.08 | tr | 98.2 | Х | Х | | Х |
| 60 | 0.05 | tr | 0.15 | 0.22 | 0.10 | nd | 59.3 | 0.03 | 39.2 | 0.68 | 0.06 | 0.03 | 86.9 | Х | Х | | Х |

All elements analysed in wt% (normalised). Approximately 10 area analyses per sample were done in an approximate area of $151 \times 88 \mu^2$ (×1000). Nd, not detected; Tr, trace element; HW, hot worked; Q, quenched; A, annealed; C, Cast.

Not detected and trace elements have been quantified as 0 in statistical analyses. Totals is the measured total (without oxygen) prior to normalization.

The high-tin content of MA60 may be a consequence of its high level of oxidation, which could have led to over-represented tin levels due to its easier oxidation.

This problem was addressed under SEM-EDS, selecting carefully only metallic areas, and tin content descended to 28.7 wt% but even so, this sample analyses should be used carefully. MA72 and 73 correspond to samples taken from the bottom of the vessels and MA72b and 73b from the rim. MA77 correspond to a sample taken from the decorated area of the handle drum and MA77b from the joint between the handle and the body vessel.



Fig. 4. Microstructure of the Vietnamese vessel. Etched. 50×, 2 mm width. Plane polarised light. Note the cored dendrites and copper sulphide inclusions in blue.

This artefact exhibits a cored dendritic structure, indicative of casting followed by fast cooling and a eutectoid inter-metallic solution composed of α and δ layers (Fig. 4). Blue segregates of copper sulphide can be seen at higher magnification (Fig. 5).

3.2. High-tin bronzes

With the exception of the ingot (MA78), all the high-tin bronzes were cast, hot worked, and quenched. The fast cooling treatment prevents the development of the brittle δ -phase and retains the β-phase that forms the characteristic martensitic structure of hightin alloys (Rajpitak, 1983; Goodway and Conklin, 1987; Scott, 1991). The knobbed vessel (MA73) and other two possible vessels (MA4 and MA66) show clear evidence of casting before hot working, with the dendritic structure still discernible despite the subsequent hot working and quenching. In MA73, the solid orange phase is the remnant of α -phase with the 'bluish' areas corresponding to the eutectoid compound of the dendritic microstructure. Hot working can be inferred in this sample because equi-axed and twinned grains can be seen inside the orange islands and the copper sulphide segregates developed a linear morphology (Fig. 6). The vessel was then quenched to produce a very hard β -phase, but one much less brittle than $\beta\mbox{-bronzes}$ slowly cooled to the $\alpha\mbox{-eutectoid}$ room temperature form (Scott, 1991).



Fig. 5. Detail of the Vietnamese vessel. Etched. 500×, 0.2 mm width. Plane polarised light. Note cored dendrites, chalcosine and the eutectoid laminar δ compound.

Remnant casting structures are less clear in MA60 (one of the indeterminate decorated pieces) and MA36, the bracelet fragments. In the latter artefact, the linear tendency of copper sulphide segregates was seen under the microscope. This morphology does not surround the surface decoration but is arranged on parallel lines, so chasing or *repoussé* does not seem to have been used as a decorative technique, but engraving may be considered likely, as supported by the lines on surface. Finally, the 'ingot' MA78 shows a columnar dendritic structure, indicative of a fast cooling from the melt, as well as copper sulphide segregates.

High-tin bronze is notoriously difficult to work at heat and impossible when cold, owing to its hardness and brittleness, characteristics described in Strabo's *Geography* when he records that "the Indians used vessels which were cast instead of beaten and which broke like pottery when dropped on the ground" (quoted in Rajpitak and Seeley, 1979, p. 28). These qualities make high-tin bronze unsuitable for tools or weapons, but the alloy's reflectivity and sonority made it desirable for bells and mirrors, just as their silver-white colour may have added appeal.

3.3. Leaded bronzes

The addition of lead can improve a copper-alloy's casting characteristics by reducing the melting point and viscosity, and increasing the workable casting range and rendering greater detail



Fig. 6. Knobbed vessel. Etched. $200 \times$, 0.5 mm width. Plane polarised light. Note the linear disposition of chalcosine and the equi-axed grains as an evidence of hot working, and the martensitic structure of the δ compound characteristic of quenching.



Fig. 7. Microstructure of the Han mirror. Etched. $500 \times$, 0.2 mm width. Plane polarised light. Note the laminar δ -phase compound. Lead appears as globular segregates.

in the mould. However, the miscibility of lead in copper is low, resulting in globular segregates of lead in copper-alloy microstructures that increase brittleness and reduce strength. Leaded bronze was used to cast three KSK artefacts: MA35 (a possible bell), MA68 (a Western Han mirror), and MA77 (the 'Dong Son' drum handle), with their lead contents ranging between 5.0 wt% and 9.4 wt%. All leaded bronzes were cast, as can be inferred from their dendritic structure, and their metallographies mainly differ on the basis of their tin contents.

The Western Han mirror metallography (MA68), with high tin levels (~25 wt%), shows clearly the hard and brittle laminar δ -phase compound. Fig. 7 shows a general view of the mirror, where α grains can be distinguished in a matrix of a eutectoid δ compound, laminar and lighter. Lead appears as globular segregates. The mechanical properties of this heavy ternary alloy are even poorer than those of unleaded high-tin bronzes (discussed above). Nevertheless, once polished, these alloys are highly reflective (Bowman, 1991; Rovira and Gómez, 2003), which is probably why they were chosen for both Chinese and Roman mirrors (Rovira and Gómez, 2003, p. 24; Scott, 1991, p. 108). Cast ternary alloy mirrors with 22-23 wt% tin have been documented from sites in the Central Plains and Xinjiang regions of China during the latter half of the 1st millennium BCE (Mei, 2000, p. 47). Guiver et al.'s (1996-1997) study of Chinese bronzes suggested that 'low-tin' bronze was the preferred alloy for weapons, tools, and vessels and leaded bronzes for coins, whilst high-tin leaded bronzes were chosen almost exclusively for mirrors. Guiver et al. (1996-1997, p. 100) also noted that pre-Han mirrors contained c. 20 wt% tin, and Han mirrors c. 25wt% tin, after which the tin content seems to have reduced, which they proposed may have been due to economic reasons.

The drum handle (MA77), also a leaded bronze, presents a different microstructure due to its different concentrations of tin (~10 wt%) and lead (~12 wt%). In particular, segregates of δ -phase appear as bright white phases (Fig. 8). This observation is in agreement with Rajpitak (1983), who states that, in practise, the brittle δ -phase appears from c. 5–7 wt% tin. Cuprite is also present, and clearly distinguished by its red internal reflections in XPL, and the cored aspect of the dendrites is indicative of a fast cooling rate. The metallography of this artefact also allowed us to determine that the handle was not soldered to the vessel but cast, as was the decoration, due to the absence of high structural stresses that are caused by chasing or engraving.

Finally, it can be noticed how the possible bell (MA35), which displayed a different chemistry, namely in its high content in



Fig. 8. Microstructure of the Drum. Etched. $500 \times$, 0.2 mm width. Plane polarised light. Note the bright laminar δ -phase compound, which has appeared despite this not being a high-tin bronze. Cuprite can also be distinguished in red.

antimony, has also different working techniques, being the only object which was annealed after casting to get a higher homogenisation through re-crystallization (Fig 9).

4. Technical ceramics and slag

Four different kinds of technical ceramics have been found in KSK: i. amorphous fragments (TC34), ii. possible pyrotechnological structures (tentatively called 'furnace' ceramics) (TC35), and two types of vessel: iii. smaller ones with a 'nipple' on the base (TC9 and TC16), and iv. a bigger one whose base is missing (TC17), so we do not know if it is a larger version of the 'nippled' type (Fig. 10). The first issue was to determine if these ceramics had been used in metallurgical activities, and secondly, to evaluate the possibility of the smaller vessels being used as ingot moulds, similar to those documented in central Thailand by Pigott and Ciarla (2007, p. 83) and the bigger one as a crucible (Fig. 11).

SEM-EDS compositional analyses of the technical ceramic fabrics allowed the differentiation of two groups, differing principally in their alumina contents (Table 3). The 'low alumina' group is constituted by the sample of vitrified, amorphous ceramic (TC34), and the 'furnace' fragment (TC35). Their structures show a highly



Fig. 9. Microstructure of MA35. Etched. 200×, 0.5 mm width. Plane polarised light. This is the only artefact that was annealed after casting to get a higher homogenisation.



bloated ceramic matrix, with abundant quartz grains and smaller and fewer inclusions of zircon, rutile and monazite. The vitrified state of these ceramics leaves little doubt that they were involved in high-temperature technologies, but given the lack of any metal traces we cannot confirm any link to metallurgical activities – they will not be discussed further.

The 'higher alumina' group comprises solely the vessels, all of which can confidently be ascribed to copper-base metallurgical activities. This might be indicative of a preferential selection of alumina-rich clays for metallurgical vessels but it should be noted that 'alumina-rich' is relative; these vessels would still have mediocre refractory properties (e.g. Freestone and Tite, 1986; Martinón-Torres and Rehren, 2009). The ceramic fabrics in this group are similar to each other, characterised by an abundance of elongated voids left by organic temper (although some organic inclusions remain) and less abundant quartz grains, zircon, and iron silicates. Another noteworthy feature of this ceramic group is the relatively low vitrification documented in the matrices, particularly for the smaller vessels. Considering the limited refractoriness that we can infer based on the ceramic composition, the lack of vitrification indicates that either they were not exposed to high temperatures or, if they did, this exposure did not last long.

Focusing now on the smaller nippled vessels, surface XRF analyses of two of them (TC9 and TC16) indicated the presence of both copper and tin, and the microanalytical study of TC9 shows traces of oxidised bronze mechanically adhering to the inner surface, with virtually no chemical interaction (Fig. 12). This, coupled with the low vitrification and their overall shape, lead us to suggest these vessels constitute ingot moulds, where the metal would solidify relatively quickly (cf. Pryce et al., 2008, p.301, who initially suggested use as crucibles). Somewhat exceptional is the 'mould' TC16, which shows a lump of bronze slag adhering to the outer surface, but this is probably owing to some accidental contact between the two. The interpretation of these vessels as moulds is consistent with the matching high-tin bronze ingot found on site (see previous section) and with the broadly comparable 'cup mould' tradition found in mainland Thailand (Pigott and Ciarla, 2007).

The larger vessel (TC17) was clearly engaged in a different process, and we interpret it as a crucible due to the 1–3 mm layer of slag adhering to its inner surface. This slag is mostly composed of fayalite crystals (Fe₂SiO₄) immersed in a matrix of K-melilite (KCaAlSi₂O₇) containing traces of tin oxide. Small prills of copper sulphide were detected in the crystalline matrix, as well as larger (~ 100 µm) prills of oxidised bronze towards the surface (Fig. 13). The oxidation of these prills is probably post-depositional, as indicated by the ghost structures preserved in all of them, together with the presence of covellite and fayalite, which indicate reducing conditions – though these were probably variable due to the coexistence of fayalite and K-melilite.

The presence of copper and tin in the crucible slag suggests four potential scenarios: i. the melting of bronze, ii. the alloying of metallic copper and metallic tin, iii. the co-smelting of copper minerals and tin minerals, and iv. the cementation of tin minerals with metallic copper.

Although some degree of chemical interaction between the slag and the ceramic fabric is noticeable at the interface, the high iron content in this slag residue, as evident in the formation of fayalite, is far in excess of the iron oxide in the ceramic (c. 3 wt%). This indicates that the charge was rich in iron and suggests that at least one of the components entered the charge in a mineral form, possibly bringing iron oxide as gangue. It is possible that this iron-rich gangue comes from panning alluvial cassiterite from 'black sands'

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Fig. 11. Top: Ingot mould from KSK with a high-tin bronze ingot inside. Bottom: mould from Central Thailand, (Pigott and Ciarla 2007, p. 83).

in a placer deposit, as cassiterite and iron-rich sands usually concentrate together in placer deposits, and prehistoric metalworkers might have been unable to separate them. However, more geological surveys should be done on this area in order to corroborate this possibility. The refining of iron-rich bronze might also be a possibility, but the relatively low iron content of the metal artefacts from KSK and elsewhere in Thailand argue against this supposition. Hence we favour the 'crucible smelting' scenarios iii and iv, instead of 'crucible melting' possibilities i and ii.

It is difficult, however, to decide between options iii and iv, mostly owing to a scarcity of comparanda (but see similar examples of bronze slags interpreted as by-products of co-smelting cassiterite and metallic copper recently found in Portugal: Figueiredo et al., 2010; Valério et al., 2009). According to Rovira (2007), slags deriving from co-smelting copper and tin minerals typically contain malayaite (CuSnSiO₅) or other copper neosilicates, as well as prills with compositions varying from pure copper to high-tin

| Table 3 | |
|--|--|
| SEM-EDS matrix analyses of technical ceramics. | |

| Matrix | Na ₂ O | MgO | Al_2O_3 | SiO ₂ | P_2O_5 | K_2O | CaO | TiO ₂ | FeO |
|------------------|-------------------|-----|-----------|------------------|----------|--------|-----|------------------|-----|
| TC9 (mould) | 0.7 | 0.2 | 19.8 | 71.5 | 1.0 | 1.4 | nd | 1.4 | 3.8 |
| TC16 (mould) | nd | 0.2 | 19.5 | 68.5 | 4.3 | 0.8 | 0.5 | 1.3 | 4.6 |
| TC17 (crucible) | nd | nd | 19.6 | 72.7 | 1.9 | 0.9 | 0.3 | 0.9 | 3.4 |
| TC34 (amorphous) | nd | 0.2 | 11.9 | 80.1 | 0.1 | 1.5 | 0.3 | 0.6 | 5.1 |
| TC35f (furnace?) | nd | 0.2 | 12.1 | 77. 7 | 0.3 | 2.0 | 0.4 | 0.4 | 6.8 |
| TC35r (furnace?) | nd | nd | 12.1 | 82.9 | nd | 0.5 | 0.1 | 0.2 | 4.1 |

All results normalised. Oxygen by stoichiometry. TC35f, furnace; TC35r, relining of TC35.

Data correspond to the average of between 7 and 10 analyses of areas between 500 and 600 μ^2 except for those areas which, due to high vitrification, had to be smaller. Nd, not detected. Not detected has been quantified as 0 in statistical analysis.



Fig. 12. Line of oxidised bronze particles at the inner surface of mould fragment TC9. Due to the lack of chemical interaction with the ceramic, bronze seem to have been mechanically attached to the surface. The mould fabric is coarse with abundant porosity (dark) and quartz inclusions (light grey), and shows only limited vitrification. SEM-BSE image.

bronzes. Such features are absent in crucible TC17. Also following Rovira (ibid.), a conspicuous characteristic of crucible slags related to cassiterite cementation with metallic copper is the presence of clusters of nodular cassiterite, which are not found here either. However, as a preliminary interpretation, we are inclined to interpret this as a crucible where bronze was made by cementation of iron-rich cassiterite with copper metal. Under reducing conditions, the bulk of the cassiterite could be reduced to the metallic state and absorbed by the copper, either by cementation or with both metals in a liquid state, depending on the temperature; whereas the same atmosphere would allow for an iron ion equilibrium towards Fe^{2+} , which can form fayalite. This tentative reconstruction is not only consistent with the analytical data, but also with the geological environment (notably, the ready availability of cassiterite and the lack of copper ores) as well as with the archaeological reconstruction (see below).

Even more difficult to interpret is an amorphous c.150 g lump, MS173, $74 \times 70 \times 40$ mm in size, originally categorised as 'bronze slag', which has both similarities and differences when compared



Fig. 13. Crucible slag (lower right half) with fayalite crystals in a potash-rich melilitic matrix and oxidised bronze prills on its surface. Dark grey are soil residues corroded onto the slag surface. Sample TC17, SEM-BSE image.

to the preceding sample. The microstructure of this piece is extremely complex and heterogeneous, alternating areas of metallic copper, oxidic slag phases and lumps of vitrified ceramic the latter, indicating that the piece may originate from a highly distorted crucible. Besides the alkali oxides that may be attributed to molten ceramic and/or fuel ash, the main compounds present in the slag matrix are oxides of copper, tin and lead. Copper is present in the form of metallic globules but also as cuprite prills, indicating a relatively oxidising system. Indeed, metallic prills are sometimes surrounded by a halo of oxidation. In other areas of the sample, copper appears bound with lead oxide and silica, forming globular phases that appear interspersed with acicular cassiterite. This phase is also very abundant, and it often appears concentrated in discrete clusters (Fig. 14 and Table 4).

Like TC17, this sample appears to be related to bronze production. The clustering of cassiterite is a strong indication that it entered the system in a mineral form, rather than resulting from the oxidation of a bronze during melting. However, in this case, the reducing conditions necessary for the production of bronze by cementation seem not to have been achieved or maintained for long enough. A further peculiarity is the presence of lead, which could have entered the system with the metallic copper or as a separate component. In the relatively oxidising environment, lead could have reacted preferentially with oxygen and silica from the ceramic, to form the phases described above. The reasons why the making of bronze was not completed in this case can only by hypothesised, although the failure of the crucible and subsequent abandonment of the operation seem plausible. The (unexpected) presence of lead, oxidised during the process, could have been the reason for the failure of the vessel in this case, as lead oxide is highly aggressive towards siliceous ceramics (Rehren, 1996)

5. Discussion

In order to understand the nature and importance of metals and metallurgy at KSK, the crucible, mould, slag and metal evidences must be brought together. The first aspect deserving attention is the stylistic variety of the metal artefacts. Their stylistic influences clearly show South Asian, Vietnamese 'Dong Son', and Western Han Chinese characteristics, which indicate the cosmopolitan character



Electron Image 1

Fig. 14. Slag droplets which often appears concentrated in clusters within the metal composed by globular copper and lead silicates (1) interspersed with acicular cassiterite (2); analysis 3 shows a general composition of this material. SEM-BSE image.

Table 4

SEM-EDS area and point analyses shown in Fig 14.

| Spectra | MgO | Al_2O_3 | SiO ₂ | K ₂ O | CaO | FeO | CuO | SnO_2 | PbO |
|---------|-----|-----------|------------------|------------------|-----|-----|------|---------|-----|
| 1 | | 7.0 | 53.0 | 1.3 | 1.2 | 1.4 | 33.3 | | 2.9 |
| 2 | | | | | | | | 100 | |
| 3 | 0.5 | 3.2 | 31.7 | | 0.6 | 1.6 | 58.9 | 1.7 | 1.8 |

Oxygen by stoichiometry. All results in weight %.

of KSK during late prehistory (Bellina, 2008). The crucial question here is whether the artefacts were imported from those areas or locally produced following foreign designs. As discussed above, it was possible to differentiate alloy types that appear correlated with the artefacts' typologies. However, the analysis of metal impurities did not allow us to discern any groupings (with MA35 as the only potential outlier). It is therefore tempting to suggest that artefacts of various types could have been made locally rather than imported, which would explain the similar chemical signature. However, it should be stressed that the pattern of impurities detected in the metal assemblage is not distinctive enough for this conclusion to be firmly established. Forthcoming lead isotope analyses, even if distorted by the heavily alloyed nature of the artefacts, may aid in resolving this question.

Turning to the rest of the assemblage, we have tentatively established the presence of nippled moulds for high-tin bronze ingots as well as larger crucibles that would have been employed for the production of copper/tin alloys via cementation of cassiterite with metallic copper. Considering the lack of copper ores in the Thai-Malay Peninsula, in contrast with the availability of tin ores (Pryce et al., 2008), the emerging picture suggest that KSK metallurgists could have imported copper or bronze from other regions (i.e. the Khao Wong Prachan Valley in central Thailand, see Pryce, 2009 or the copper mining complex at Phu Lon, Pigott et al., 1992), and 'added value' by cassiterite cementation. The resulting high-tin bronze could have been employed locally for artefacts such as those analysed here, but the presence of ingot moulds and a hightin bronze ingot strongly suggest that production was also oriented for exchange. It has to be mentioned that high-tin bronze artefacts have also been documented at Ban Chiang, Don Klang and Ban Na Di in Northeastern Thailand, and Ban Don Ta Phet in Western Thailand (see Pigott et al., 1992). Another question that should be discussed further is the possibility of high-tin bronze technology being spread from Southeast Asia to the western Islamic world, as high-tin bronzes were the most important copper-tin alloy used in the Islamic Iran and its introduction is still under discussion. The technology could have come from China (as high-tin bronze mirrors similar to Chinese ones have been found there) but it could have come also from Southeast Asia, as copper mines were common in Iran but tin ores were not (Pigott and Allan, 2009).

Although we have compared KSK's technical ceramics to others elsewhere in Thailand, there has been no convincing analogy for the particularly distinctive nippled wares. However, strikingly similar ceramics have recently been reported along with a high-tin bronze ingot from the broadly contemporary site of Tilpi in West Bengal (Datta et al., 2007). Given that high-tin bronzes probably date from at least the early 1st millennium BCE in South Asia (Srinivasan, 1997, 1998, 1999; Srinivasan and Glover, 1995), it is conceivable that KSK high-tin bronze production technologies were transmitted by some means across the Bay of Bengal. Such a possibility is in accordance with Bellina's original (2001) model of South Asian artisans settling at KSK and using South Asian techniques to produce 'Southeast Asian' material culture (Bellina, 2003, 2007, 2008). However, it must be emphasised that KSK's metallurgical and glass industries lack the rigorously quantitative skillbased ethnoarchaeological studies (Roux, 2000; Roux et al., 1995)

that structure the hardstone ornament artisan 'limited migration' hypothesis.

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6. Conclusion

Bearing in mind that KSK has now been almost entirely obliterated by looting, the relatively low sample numbers of the present study permit a tentative reconstruction of late 1st millennium BCE copper-base metallurgical activities in the Upper Thai-Malay Peninsula. Here are our concluding points.

- According to the trace element similarity of the artefacts, local production of the assemblage following foreign styles seems the most plausible hypothesis, although lead isotope analyses should provide useful insights.
- Technical ceramics can be classified due to their functionality into ingot moulds and crucibles, although more analytical work has to be done in order to contrast this proposition.
- 3. Archaeometallurgical by-products also suggest local high-tin bronze production, perhaps in relation to the contemporary industrial-scale copper extraction in the Khao Wong Prachan Valley. Current evidence suggests the possibility of cassiterite cementation at KSK although more analyses should be done in order to confirm this hypothesis.
- 4. Both the working techniques exhibited in this artefacts assemblage, as well as the bronze production by cementation shows the advanced skills of KSK metalworkers and their knowledge of alloying effects and working properties.

If our reconstruction even partly coincides with historical reality, not only would KSK have provided the earliest evidence for the exploitation of Peninsula tin in the last centuries BCE and confirmed a long suspected material contribution from the Peninsula to the trans-Asiatic exchange network, but it would also support Bellina's (2001) original thesis for highly skilled South Asian artisans having settled in Southeast Asia or trained Southeast Asians – whether at the behest of local social groups or of their own volition. The archaeometallurgical research conducted by the Khao Sam Kaeo Archaeological Mission has now yielded further multi-disciplinary evidence for the prehistoric meeting of South, East, and Southeast Asian social groups, with KSK itself acting as a major cosmopolitan trans-Asiatic industrial and exchange centre participating in the circulation of skilled technologies and hybridised cultural products.

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