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Research on copper production of the Bohai Kingdom in Northeast China

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Abstract

The systematic investment and rearrangement of the unearthed artefacts of Longtoushan Royal Cemetery of the Bohai Kingdom have provided a bunch of new information for the archaeometallurgy research on non-ferrous metal production. We have synthesised the latest discoveries of several related mining/smelting sites and urban relics for making a direct approach to the chain of copper production in this research. SEM-EDS and p-XRF unveiled the distinction between copper and bronze artefacts. The yield gap between the scale of smelting and the number of artefacts was also discussed based on historical records. The evidence and detail of indigenous copper production could help the researcher comprehend the technology and material exchange and geopolitical relationship development of the Bohai Kingdom.

Keywords: Bohai Kingdom, Tang Dynasty, Bronze, Copper, Archaeometallurgy

Introduction

At the northern Frontier of the Tang Dynasty, Bohai Kingdom was one of the early kingdoms founded by the Sumo Tribe of Mohe People [1]. The Bohai Kingdom lasted over two centuries (698–926 AD) prosperously in the autonomous administrative and political organisation system of the Tang Dynasty ('Jimi' system). This research was based on a task for collating and arranging excavation data of the Longtoushan Tombs' (meaning Dragon Head Hill and abbreviated to LTS) latest excavations. The LTS were intermittently excavated since 1980. The unearthed tombs with inscriptions and symbolic ornaments, such as gold crowns and high-class construction components, have suggested the area as the royal cemetery of the Bohai Kingdom [2]. The Jilin Provincial Institute of Cultural Relics and Archaeology, cooperating with other academic institutions, carried out a series of archaeological excavations from 1997 to 2008. There were hundreds of non-ferrous artefacts unearthed during the latest excavations, including exquisite artware and small pieces of ornaments. The copper and bronze artefacts were the most abundant unearthed metal relics, which could be valuable for investigating the craft production system. The distinction between the indigenous and imported artefacts was also the precondition for elucidating the mechanisms of distribution of materials [3].

The Bohai Kingdom records no official history, and other literature about it from Tang, Khitan (Liao), Silla and Japan was also limited. So, archaeological researches were the most reliable source. Most of the Bohai sites are located in China and North Korea, and Russian scholars have also studied Bohai (Balhae) for more than 151 years with systematic excavation at their eastern frontier (Fig. 1) [4]. Hundreds of copper and bronze artefacts were unearthed from Bohai sites. A series of copper mining and smelting sites in the Bohai Kingdom were discovered recently [5], which suggested local mining and smelting activities for producing copper. Since the bureau document of the Tang Dynasty has recorded an official application on the maintaining the trading of blister copper (recording as 'Shu Tong') with the Bohai Kingdom, the copper material could be essential in the material distribution between Tang and Bohai [6]. Still, according

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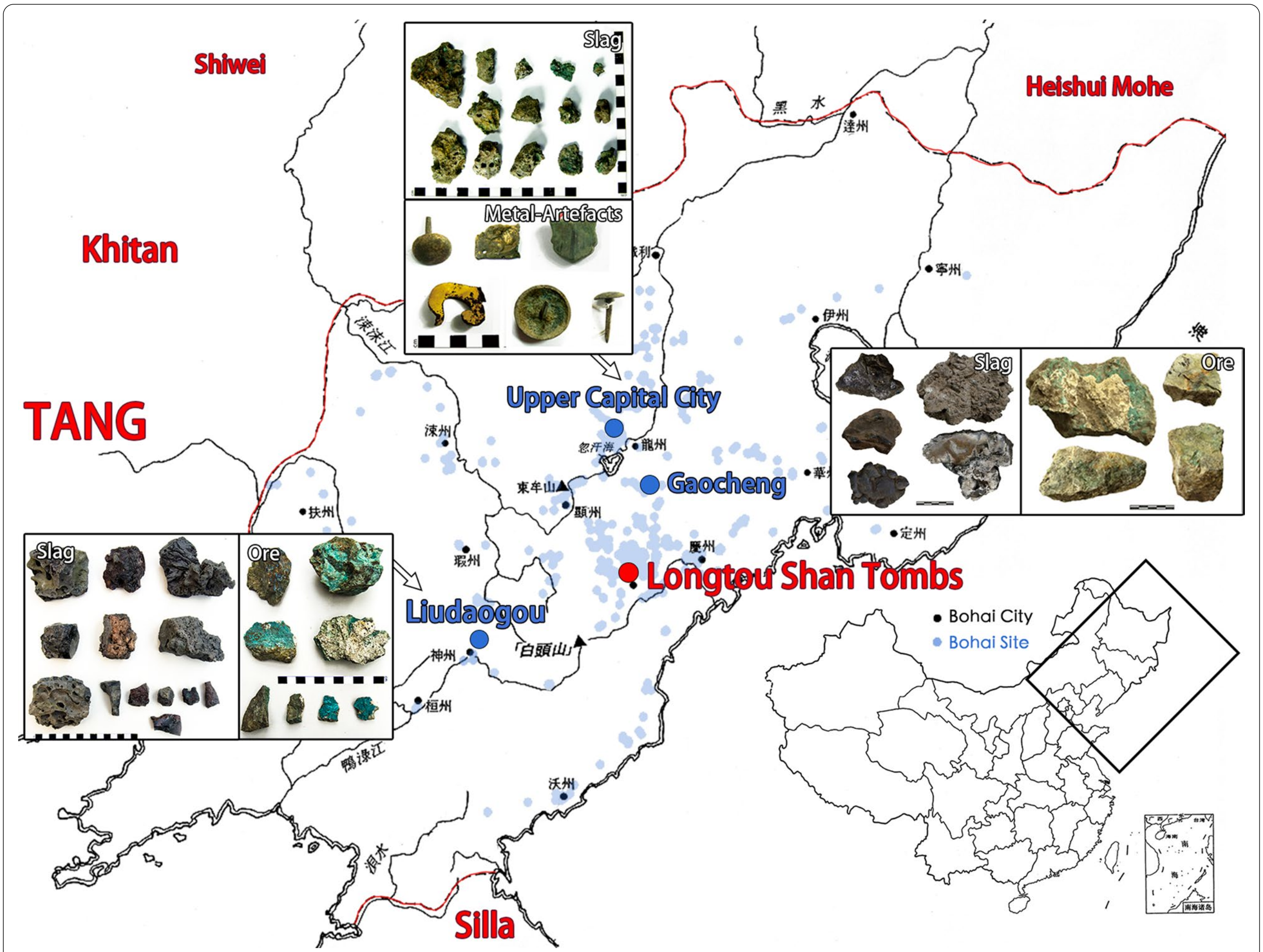


Fig. 1 The Bohai Kingdom sites distribution and relic location

to other documentary records, tributaries between the Bohai Kingdom and Tang Dynasty only included fur, herb, gems, and horses for silk and artware [7]. So this research tried to work through the relics of Bohai from a perspective of copper production for further comprehension of the society of the Bohai Kingdom.

Sites and research background

For such a purpose, some of the tombs of LTS were selected to review the non-ferrous relics of the elite class. To the southeast of the capital city of the Bohai Kingdom, LTS lay at the core region of the Bohai Kingdom. The whole excavated area was divided into two zones, Longhai (meaning Dragon Sea, abbreviated to LH below) and Shiguo (meaning Stone Realm, abbreviated SG below), by the distribution of more than 20 tombs and other relics on the landform. The selected six tombs were the most preserved ones (most of the tombs were unfortunately robbed), and 271 pieces of metal artefacts were sampled based on the latest organised excavated units [8]. Four tombs belong to the SG area (M1B, M1C, M2A, M2B) and two tombs to the LH area (M10, M13) therein. For comparing purposes, more bronze artefacts unearthed from the commoners' tombs in eastern Jilin province and southern Heilongjiang province were involved in this research (Fig. 1).

To complete the whole craft production system of copper, archaeological reconnaissance was conducted in the two modern polymetallic copper mining regions in the Bohai realm. Two mining and smelting sites were discovered and excavated (Fig. 1), which were the Baoshan-Liudaogou site group at Linjiang City (abbreviated to LDG below) and the Gaocheng site at Wangqing County (abbreviated to GC below). Hundreds of copper ores and smelting slags were excavated, and over 50 pieces were sampled for this investigation [5, 9]. LDG contained one ancient mining zone, four smelting sites, several small-scale dwelling sites, and a docking site. Combining with the sites, the region indicated a series of raw copper material production activities in the Bohai Kingdom. The excavation circumstance of GC was still confidential, the excavator provided only ores and slag samples.

The residual remains of craft proceedings were scarce; only one was found at the Shangjing Cheng site (meaning Upper Capital City & abbreviated to SJ below). SJ was the ruin of the essential city of Bohai Kingdom, which was regarded as the kingdom's capital for over eighty years [10]. 75 pieces of the relics sampled in this research were unearthed from four excavation zones: the Palace, the Eastern Gallery, the Workshop, and the Royal Garden. The samples included metal ornament, crucible pieces, and alloying/refining slag [11]. Since most of the artefacts collected in the museum were not allow for destructive sampling, and the preservation status of the relics was quite different, a mixture of two different detecting and sampling strategies were adopted for the specimens.

The non-destructive investigation is mainly tested with portable XRF and microscopes. Observing the well-preserved samples with minor corrosion on the surface with the KEYENCE VHS-900 ultra-depth microscope could provide technical details on the surface. And the NITON series portable XRF detection equipment XL3t-T2-500 from THERMO would also give reliable data on the artefacts' surface even with several micrometres of corrosion covering or gild golden layer [12]. But we also can not neglect the chemical processes like decuprification or other severe corrosion diseases affection for the element composition on the surface [13]. So, all the samples in this research have undergone the same standard testing process with the same p-XRF equipment in the Alloy Mode without standard and the toggle aiming; the testing durations were all set at 30 ± 5 s. The surface element composition results of the artefacts were compared with the SEM-EDS data in the section on several destructive specimens for quality checking.

These restrained destructive detections have been performed on some wreck unearthed specimens with thick rust on the surface, which would affect the result of non-destructive testing. These samples were cut and embedded with TROJAN resin and polished at room temperature for metallographic analysis with the DM400M microscope of LEICA. In addition, to obtain a more accurate composition and morphology characterisation, the polished specimen was coated with carbon film spraying treatment for SEM-EDS investigation with TESCAN's tungsten filament light source VAGA3-XMU scanning electron microscope and BRUKER's FLASH-DETECTOR series 610 M energy spectrum analyser. The EDS test was set as a non-standard with phi-rho-Z mode for 30 s valid time on each point.

The comparison with the p-XRF data and electron microscopy energy spectrum data of the identical specimens found that the XRF data of the object's surface consisted of more mercury, zinc, and iron than the energy spectrum results of the matrix. The SEM-EDS investigation showed that the residual mercury element was only found mainly residual at the interface under the gilt layer, unlike the other alloying elements primarily present in the bulk material. Zinc and iron data were highly related to the surface attachment and rust layer, barely found in the bulk material. So, these three elements were not regarded as the object of metal constitution analysis. To reduce the influence of the gilt and rust layer, we had taken the ratio of alloying element to copper element as the analysis object. The data can provide the quantity relations on the proportion of the bronze bulk material more intuitively than the weight%, which is convenient for data comparison and analysis. And the p-XRF and SEM-EDS data of lead, arsenic, tin, bismuth, and antimony were agreed well through this data processing, but only for qualitative, not quantitative research.

Results

There were 271 valid samples of non-ferrous metal specimens analysed in LTS, and the number of gilt-gold copper ornaments is about 72%. The statistical result has shown that the type of them are mainly coffin rings, bubble nails, buckles and decoration sheets (see Table 1). Considering the surface corrosion and decuprification proceeding affection, we set the weight% at 5% of Pb and 1% of Sn, As and other alloy elements as a line to distinguish the copper and bronze artefacts with the p-XRF data and 1% of all the alloy elements with the SEM-EDS data. The result was also shown in Table 1. The data distribution shows a similar pattern of non-ferrous specimens unearthed from the Shiguo Tomb and Longhai Tomb areas. The same kind of copper and bronze artefacts barely coexisted in the same tomb, and the bronze ones were absent in LH but abundant in SG. The p-XRF data show that the chemical composition of bronze is mostly copper alloy containing arsenic, tin, and lead. Most of the specimens contain trace amounts of bismuth and antimony. And the copper ornaments were relatively pure, and only lead and silver could be widely detected in the bulk material. Based on the p-XRF detection data of LTS specimens, we conducted a statistical pattern on the bronze and copper relics. The data of zinc and iron were also involved as an indicator of the corrosion condition of the specimens (Fig. 2 upper left). Since these two elements were not detected in the metal bulk of specimens by SEM-EDS, this pattern was set as a baseline for comparing with other data clusters to estimate the relationship base on the element composition and shown in the background of each coordinate system translucently in Fig. 2.

Table 1 The Samples Taken in LTS and Material Info. Statistics

Specimen type	Tomb unit	p-XRF	SEM-EDS	Total	Copper	Bronze
Bubble Nail and Coffin Ring	SG-M1B	59	6	65	1	64
	SG-M1C	0	26	26	26	0
	SG-M2A	44	0	44	0	44
	SG-M2B	0	2	2	0	2
	LH-M10	11	1	12	12	0
	LH-M13	31	14	45	45	0
Buckle and Decoration Sheet	SG-M1B	3	1	4	1	3
	SG-M1C	13	2	15	0	15
	SG-M2A	18	4	22	0	22
	SG-M2B	0	0	0	0	0
	LH-M10	28	3	31	31	0
	LH-M13	4	1	5	5	0
Summation		211	60	271	121	150

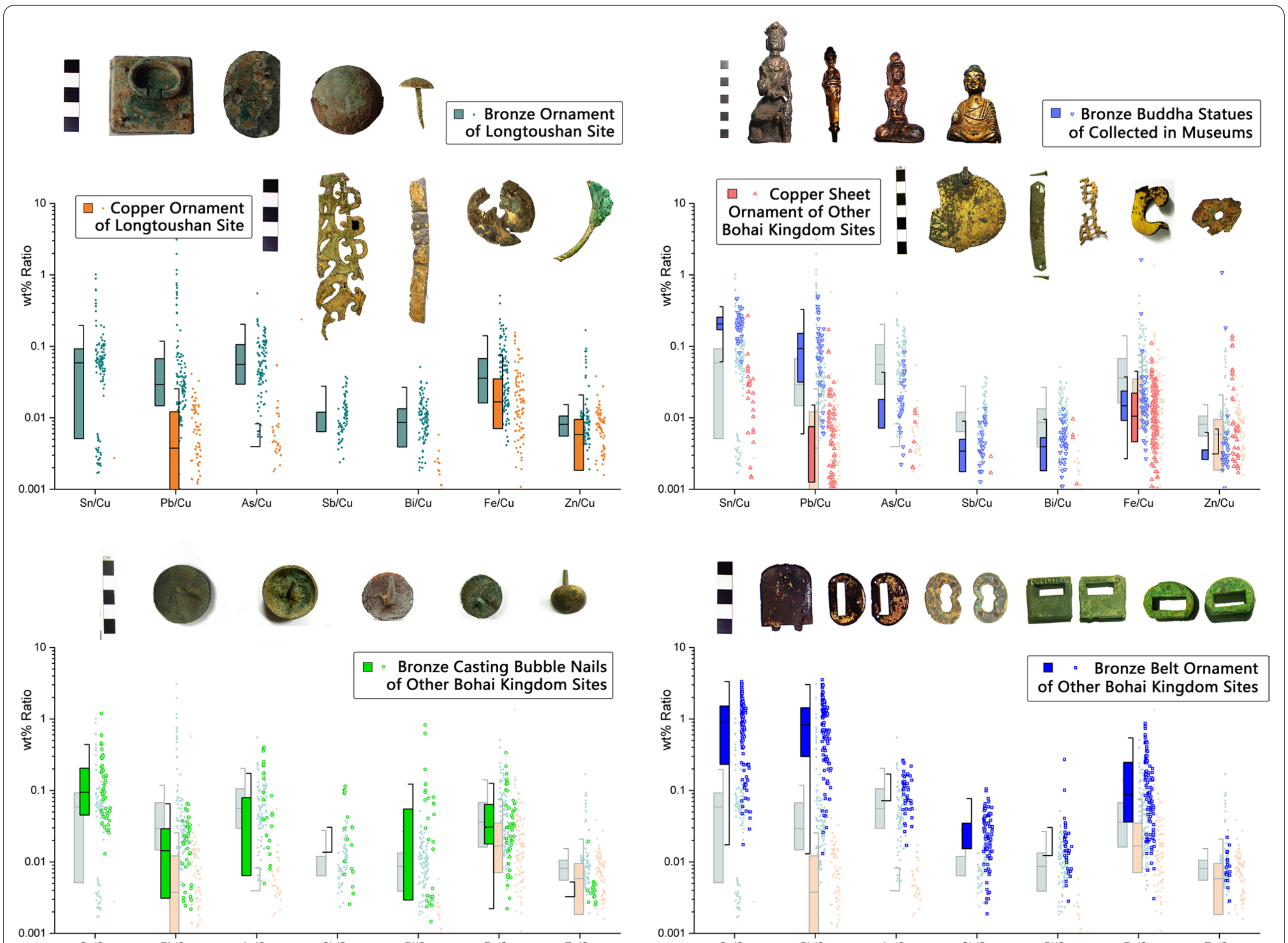


Fig. 2 The elements constitution patterns of Bohai kingdom copper/bronze artefact

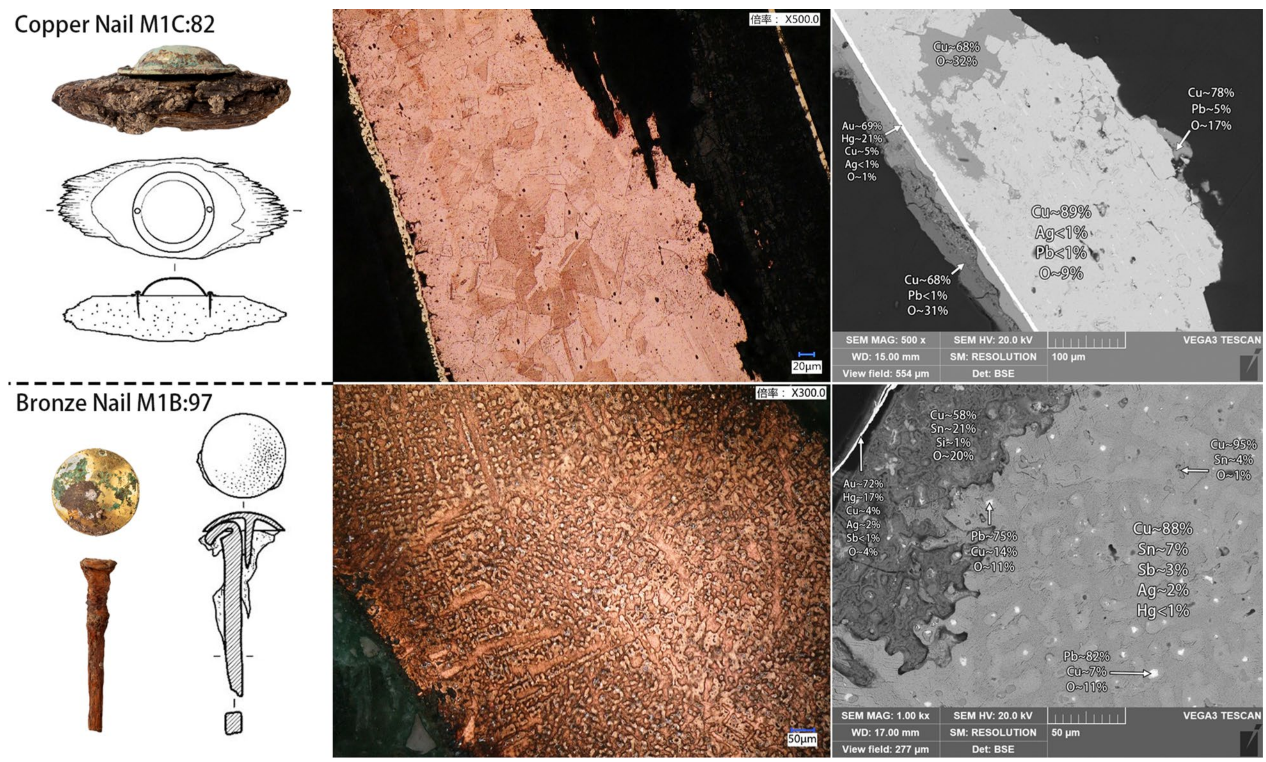


Fig. 3 The metallographic and SEM-EDS figures of copper and bronze artefact

Since LTS is considered a royal cemetery, we selected some gilt artefacts from the urban capital region of the kingdom as references to the ones that belong to the elite class. Such as Bohai-style gilt bronze Buddha statues and gilt copper ornaments unearthed from the SJ Palace and Royal Garden for the p-XRF test[1]. The 'numerical distribution' of the composition data of the two types of relics in each element component has shown a somewhat similar pattern to the artefacts of LTS. Only the tin portion of the bronze Buddha was concentrated at a relatively high level (Fig. 2 upper right). Involving the commoners' tombs from a border perspective on all the Bohai sites, we selected the bubble nail and buckles as targets, the two most widespread kinds of unearthed ornaments[2]. The alloy composition distribution of the bronze bubble nail agreed with the bronze data of LTS well, and there is no batch of copper bubble nails found at these commoners' sites in the Bohai Kingdom (Fig. 2 lower left). Buckles were also made from bronze, and the tin and lead content were higher than the bronze baseline of the LTS artefact (Fig. 2 lower right). All the data with a zinc or iron ratio higher than one was abandoned, considering the rusty surface would affect the p-XRF data too much in this research.

The metallographic phase observation and SEM-EDS investigation of interventional sampling of LTS show that the process of making copper and bronze artefact was quite different. A hot forging process mainly produces the copper relics unearthed at M10 and M13 with pure copper. The metallographs showed the residual twin crystalline phase and elongated inclusion in the bulk material, which referred to the large deformation and heating treatment during the production. Similar techniques are found in the copper bubble nails unearthed at M1C of SG and other copper decoration sheets in SJ (upper Fig. 3). All the bronze artefacts of LTS were produced by casting. The ($\alpha+\delta$) eutectoid structure formed dendritic segregation shown in the bronze specimens' metallographic figures. And the segregated structure has no deformation with relatively rare copper sulfide inclusions (lower Fig. 3). Among the 60 samples for interventional testing. However, the raw material of the artefacts was different, and their gold gilt layer shared a resemblance in both the structure and content. The thickness of the gilt layer of

[1] The buddha statues and decoration sheets specimens were provided by the Upper Capital Ruin Museum and the Archaeology Bureau of Heilongjiang Province.

[2] The bronze buckles and casting bubble nails unearthed from Xincheng site, Beida tombs, Dongqing tombs, Hongzunyu tombs, Liudingshan tombs and collections of Baishan City Museum, Fusong City Museum and Archaeology Bureau of Jilin Province.

Fig. 4 Surface Emblazon details of gold, silver, copper, and bronze artefact

the small copper and bronze ornaments was around 1–3 micrometres (Fig. 3), except for two crown decorations, which were covered by over 6-micrometre-thick golden layers. The content of the gold layer was a mixture of gold, 1–5% silver, and 10–23% mercury, which are the typical constitution of the gold amalgam for fire gilding. The SEM-EDS detected the copper specimens containing slight lead and a trace amount of silver. The bronze specimens mainly bore insoluble lead within the tin/arsenic eutectoid segregation with a bit of antimony bismuth conformed to the p-XRF data (Additional file 1).

The mechanical characteristics of copper and bronze are somewhat different and directly affect the processing of the artefact production, even for the same type of ornament like bubble nails. The copper nail was forged into a hat-like formation and fixed on the wooden coffin surface with two tiny copper nails on the rim (upper Fig. 3). The bronze bubble nails were just like the large pushpin. They were nailed to the coffin (lower Fig. 3). Both kinds of bubbles were plain surfaces with 2–4 centimetres diameters and used to fix the fabric onto the surface of coffins for decoration. The different techniques and materials combinations of the same product could cause by distinct technology traditions. The ultra-depth microscope observation provided some clues by the details of the emblazonry. The themes, types, and lines of the patterns on the gilt copper coffin ring (M13:27) were carved on the surface, which was quite similar to the silver-back of the bronze mirror (M13:168) and the golden crown (M14:1) unearthed in M14 of the LH [8]. The fluency lines and roe-like pattern carved by hollow chisel constructed the basic elements of the emblazon background. Similar carving techniques could be found only on the copper artefact, such as the leaf-shaped gilt-bronze crown ornament (M1C: 12) unearthed in M1C of SG and other decoration sheets. On the contrary, the surface treatments of bronze artefacts were quite different. The gilt bronze coffin ring (M2A:75) unearthed in the M2A of SG with entangled floral patterns was combined with little carved dens by a needle-like chisel. The techniques of the bronze process may be related to the difference in the mechanical properties of the material (Fig. 4). The diameters of the two kinds of coffin ring heads were 11–12 centimetres, and their thickness was around 2–3 millimetres. Though the size and type were rather similar, the carving techniques of copper ones were much

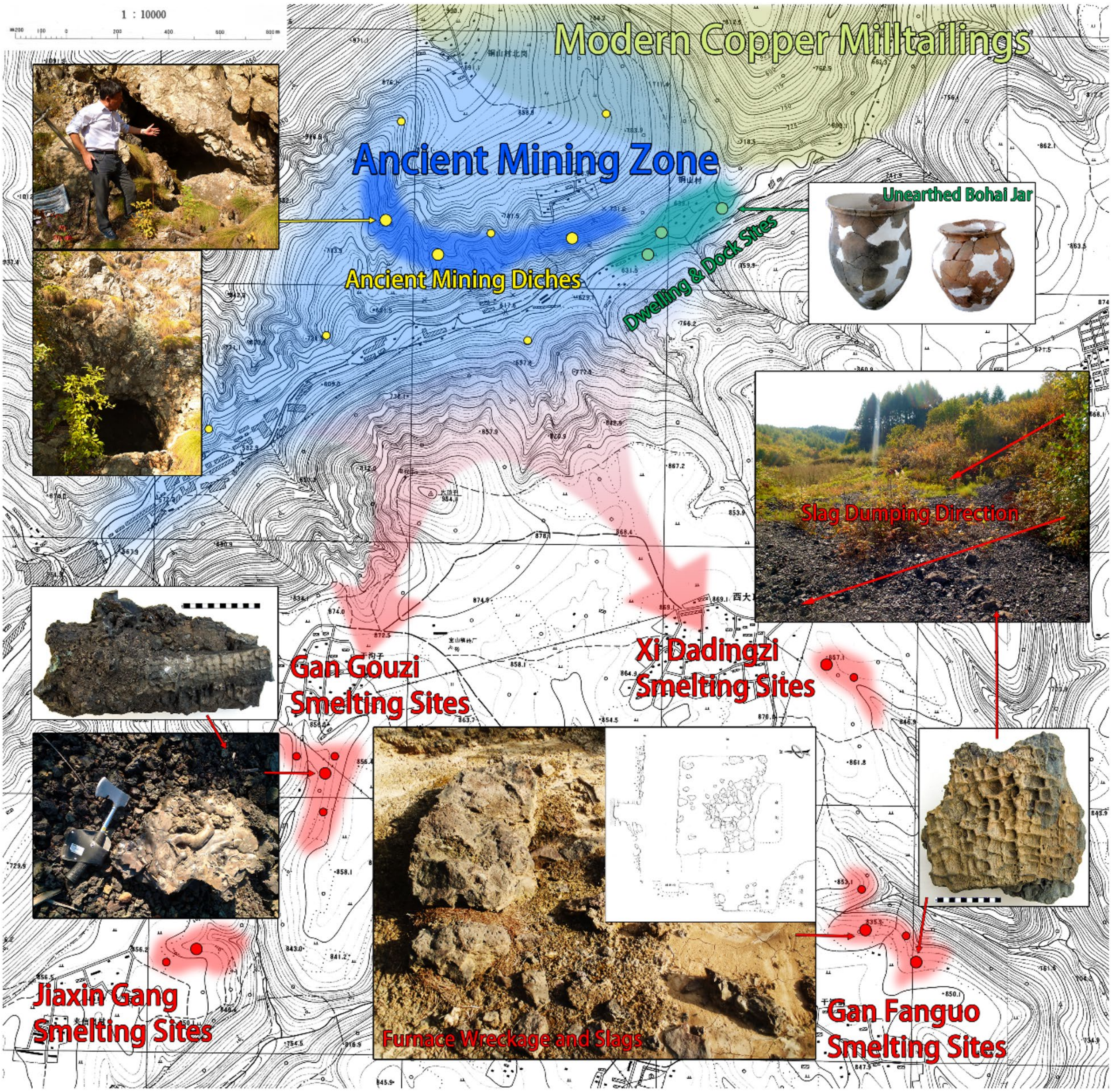


Fig. 5 The relic and site distribution of Liudaogou Site Group

more complicated and sophisticated. And the production details of the artefact would need more processing relics for further study.

To seek the provenance of the raw materials of these artefacts, archaeological reconnaissance of LDG was conducted in 2014 and 2016 on LDG and GC sites, the essential mining and smelting sites discovered in the Bohai Kingdom. LDG had been excavated about 20 years ago, and the field investigation and sampling of the specimens were mainly based on the former archaeology records. The figure of LDG was made based on a 1970 s map of this zone because modern mining production has already changed the landscape. The records of GC can not be reached before the publication of the archaeology report, and the site has been backfilled after the excavation. Fortunately, we had been authorised to test some of the unearthed slag and ores as a reference.

According to the records of the excavation of LDG, there were more than twenty ancient copper mining ditches discovered in the mining zone of LDG (see the

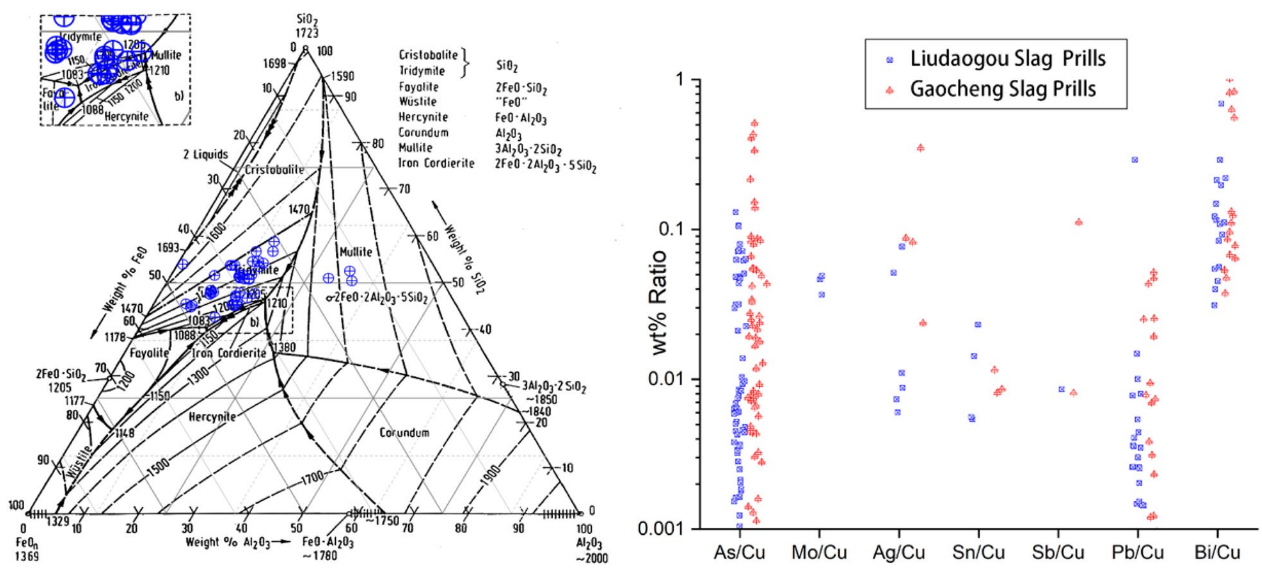


Fig. 6 The SEM-EDS data of LDG smelting slag (L) and the content of the big metal Prill embraced in smelting slag (R)

mining ditches in Fig. 5). The ditches were contacted by very narrow tunnels which could barely accommodate a single miner. Plenty of greenish ores could still be found near the ancient ditches now, which were all polymetallic copper ores (upper left of Fig. 7). Pyrite was the primary copper mineral of the skarn ores. Blending with the pyrite phases, molybdenite, bismuthinite, and stibnite could be found as the paragenetic minerals in the ores of LDG. More andradite and tetrahedrite minerals were discovered in the GC ores than in LDG; besides, the ores' content from both sites was quite similar [5]. As the former excavation and latest reconnaissance records, the living relics like the Bohai Kingdom ceramic fragments were few in the ancient mining zone. Most of them were distributed along the valley where the dwelling and dock sites were excavated (green zone in Fig. 5). Unfortunately, it was impossible to estimate the scale of mining activities since part of the ancient mining and dwelling zones were buried by modern copper mill tailings. However, about 5–10 km to the south of the mining zone, there were still several piles of smelting slag well preserved in at least four sites.

The Gan Gouzi (means dry valley, abbreviated to GGZ), Xi Dadingzi, Jiaxin Gang, and Gan Fanguo (means rice pot, abbreviated to GFG) were believed as four major Bohai smelting sites at LDG. The sampling of smelting slag at GGZ and GFG, because these two sites were better well-preserved and located far from modern living zone. Tons of the smelting slag piles at GGZ were dumped down from the top of the hills and accumulated over one meter thick on the hillside (right of Fig. 5). The slag of GFG was also accumulated around the site and at the top of the hill. We could still find the excavated wreckage of the smelting furnace at GFG, which was built up with stones nearby and clay. This copper smelting furnace was similar to the typical iron blast furnace of the Bohai Kingdom [14], with a smaller diameter of around 5 m. Although there was no trace of smelting found at the site, the indentation on the slag around the furnace had been stamped from wood charcoal patterns. These patterns could be widely found on most slag piles identified as pines log charcoal, providing relatively high calorific value. And this kind of pines is still the primary local ligneous plant nowadays. According to the former study on smelting slag [9], with adequate high-quality fuel and proper furnace design, most slag with similar content could reach a reasonable softening, and melting temperature around 1200–1400 °C [15], Some abnormal composition of the slag might cause by the erosion of adhering furnace parts (Fig. 6).

The previous research on the little metal prill, including the slag matrix, suggested that the final production of the smelting was mainly high-grade matte and blister [9], which contains primarily arsenic, lead, and bismuth. And in a little of the matte and bronze prill, antimony, tin, and silver could also be found as trace compositions. All these elements could be found in the ore copper mineral collected at LDG and GC. The ore from GC was similar to the typical poly-metallic ones of LDG, but the slag matrix constitution was relatively dispersive. Since we do not have the excavation record of the GC slag, the smelting processing details could not be deduced accurately. What

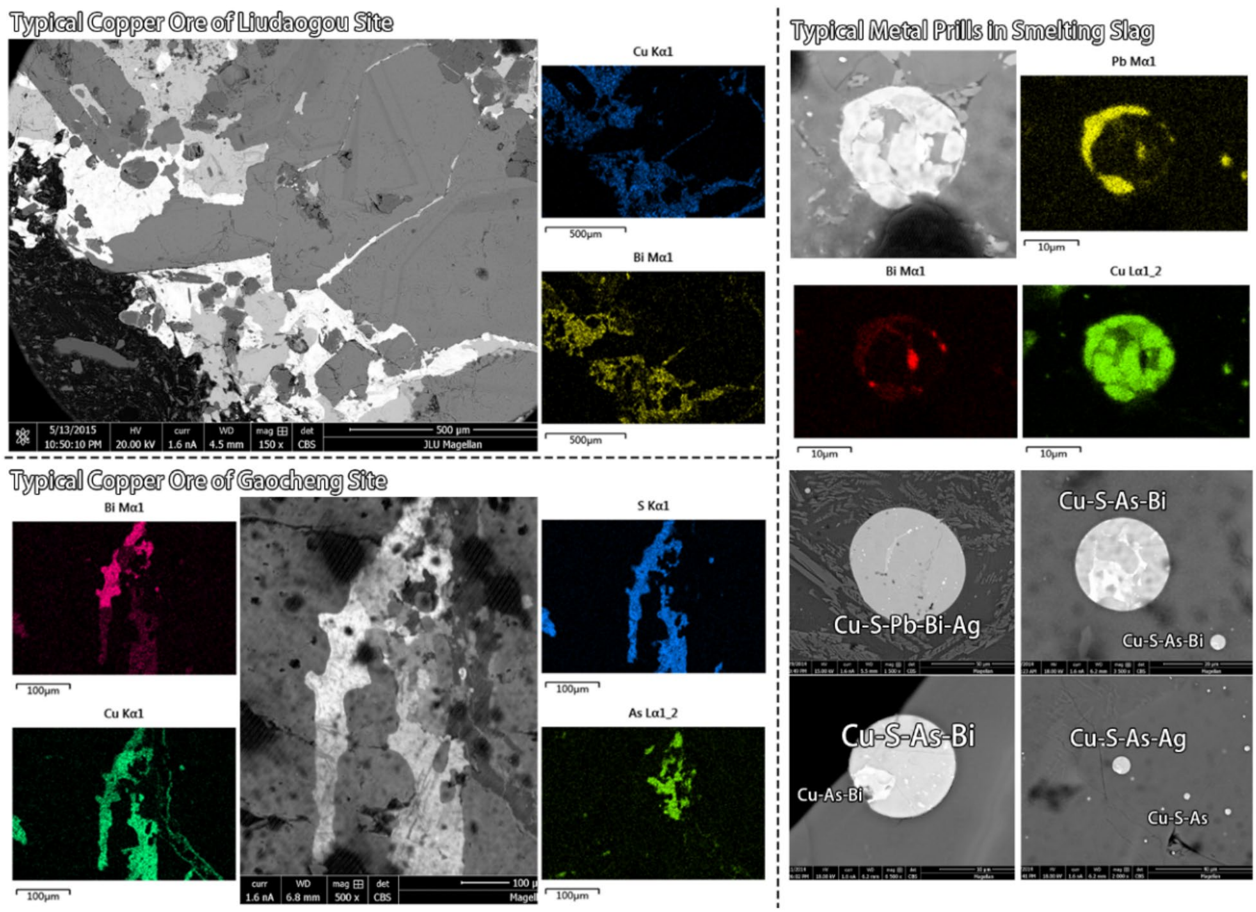


Fig. 7 The SEM-EDS figures of slag and ore unearthed from Liudaogou and Gaocheng Sites

we can confirm from the relics of GC were the production and fuel. The vegetation of GC was quite the same as LDS, and a similar charcoal pattern could also be found on the slag (the left image of Fig. 5). The whole processing of the smelting activities, which was deduced from the evidence we obtained for now, of GC and LDG should be similar to each other. The distinction in the slag content could be relevant to the different minerals within the copper ores (Fig. 7). There were records about the content data of Bohai copper artefacts unearthed in Eastern Siberian in Russia, which also contains tin, arsenic, and antimony [16]. The bubble nails and buckles were also unearthed from the Bohai sites like Kraskino on the northern shore of Russia and the northeastern Korean Peninsula, which shared the same typological characteristic of bronze artefacts [17].

Discussion

The artefacts unearthed from the tombs and the blister smelting from the ore provided information to deduce the production chain from the beginning and the end. However, the whole system still needs more process details to be complete [18]. Combining this archaeological evidence about copper mining and smelting with the test result of copper and bronze artefacts in the Bohai Kingdom, we could primarily divide these non-ferrous metal artefacts into two categories. The copper specimens are represented by the ornaments unearthed in M1C, M10, and M13, with a relatively pure element composition of these relics than the others (Fig. 2). Besides the copper specimens we have tested, other copper artefacts, including the floriated ornaments of the tomb of Princess Zhenxiao, gild sheet ornament pieces of Eastern Corridor in SJ, the coloured ornaments, earrings of Beida cemetery, were mainly unearthed from the sites around the core zone of the Bohai Kingdom [5]. On the other hand, the different type of artefact was cast with limited surface decoration from bronze alloying tin and lead and minor components like arsenic, antimony, and bismuth. These bronze artefacts were discovered not only in SG but also widely unearthed in the commoners' tombs and sites of the Bohai Kingdom. Similar bronze bubble nail and buckle specimens of Bohai Kingdom were collected from Yanbian Korean Autonomous Prefecture Museum, Fusong City Museum, Baishan City Museum, and several large-scale cemeteries like Liudingshan and rainbow trout farm sites [19, 20] were all shared analogous alloy content and typological style (Fig. 2).

There was no record of the craft production in the Bohai Kingdom, and the only reliable evidence about the craft proceeding was the slag and crucible fragments unearthed from the workshop site in SJ. The SEM-EDS data of them provided some details of the bronze artefact processing. The previous research studied the matrix constitution and inclusion distribution, and the slag was identified as tin bronze alloying/remelting slag [21]. The slag could be dumped from different batches of the alloying/remelting proceeding according to the different content of bronze prill inclusions (tables in Fig. 9). Most of the average content of the metal inclusion was similar to the bronze artefacts unearthed from LTS and other sites, which also contained tin, lead, arsenic, antimony, and bismuth (Upper in Fig. 9). Only the tin bronze prill of slag4 was relatively pure without any other alloying elements. Copper artefacts were also unearthed at the same site in SJ (Lower in Fig. 9). In fact, the individual contents of the metal prill of slag were relatively discrepant [11], which could cause by the raw material of the alloying process. There were tin bronze, arsenic bronze, lead-tin bronze, and even pure copper prill as inclusion in the same slag, which might indicate multiple copper raw material resources or materials recycling at the workshop.

Lead, tin and arsenic were all common alloy elements to enhance the mechanical properties of bronze artefacts which could all be found in the slag of SJ. But cassiterite was the only precursor mineral found in the alloying slag (Fig. 9). Comparing the content data between the smelting prill (Fig. 6) and artefacts, the arsenic should not be deliberately added to the bronze. And the three minor alloy elements, arsenic, antimony and bismuth, seem highly related to each other in our qualitative statistic on the mass ratio to copper by p-XRF(Fig. 8). The fluctuance of the arsenic composition could relate to the thermostability of itself and mixture of the raw material. As the discoveries at LDG and GC, the copper raw material supply was developed on the relatively adequate poly-metallic ore, and these paragenetic minerals could be smelted as alloy agents that could provide higher velocity in casting and better mechanical strength, like durability and stiffness, than the forging copper one. Considering the natural resource of copper with tin, arsenic, antimony, etc., and the similar cast bronze pushpin-style bubble nail and buckle ornaments unearthed since the bronze age in this region [22], we could identify these bronze artefacts production proceedings as indigenous manufactured. This casting bronze technology package could provide a straightforward processing and higher efficiency for monotonous production like bubble nails and buckles. As the evidence was shown with the archaeometallurgy specimens from the workshop site of SJ, the bronze artefact could be cast and alloyed by the Bohai craftsman from raw materials like the blister copper from LDS and GC. But the source of tin and lead would still need more trace for completing the supply system.

It is not difficult to see the difference between the copper and bronze artefacts and to deduce that the delicate pure copper ones were imported, considering only polymetallic copper ore veins within the Bohai borders [23]. In LTS, most of these copper ornaments were buried with noble metal lacquer box (M13:160) and silver back bronze mirror (M13:168) together [8], which were considered from Tang Dynasty [24]. In addition, the pattern theme and emblazonry design on the copper, gold, and silver artefacts were similar to the ones on contemporaneous Tang style noble metal artefacts and high classes bronze mirrors (Fig. 4), which indicated the copper ornaments were highly valued and relevant to the tributary or merchandise activities with Tong Dynasty. The 'sancai' multi-colour glazed maid figurines in Tang styles were considered representative relics indicating that communication with Tang Dynasty. They were also highly related to the copper artefacts, but the provenance data was not all from the kilns of central plain [25]. The artisans of Bohai Kingdom could master part of the techniques of the craft production and absorbed the design of the import artefacts for the indigenous manufacture, which not only happened on the ceramic [26] but also metal production(Fig. 4). Although copper is softer than bronze, it has better ductility for post-processing, like carving and forging, so all the floral and cirrus pattern sheet ornaments were made from copper. Moreover, the copper and bronze bubble nails in different sizes were the most decorated parts of the coffins, and the weight of the copper ones was around 70% less than the bronze ones with the same diameters with less thickness.

The isotope investigation was the most popular for the provenance research [27], but in this study, the embraced prill in the processing slag has reflected the remelting and remixing/recycling of different copper raw materials. This scenario was not commonly supported by archaeological discoveries in the bronze age but could not be continually neglected in the iron age [28]. And what is noteworthy is that some of the gilt artefacts contained a pretty high level of silver (lower right Fig. 9). Since the mechanical and colouration properties would not be affected by deliberately doping this amount of silver, the irregular high silver portion could also cause by the material recycling of copper artefacts with silver. Recyclable

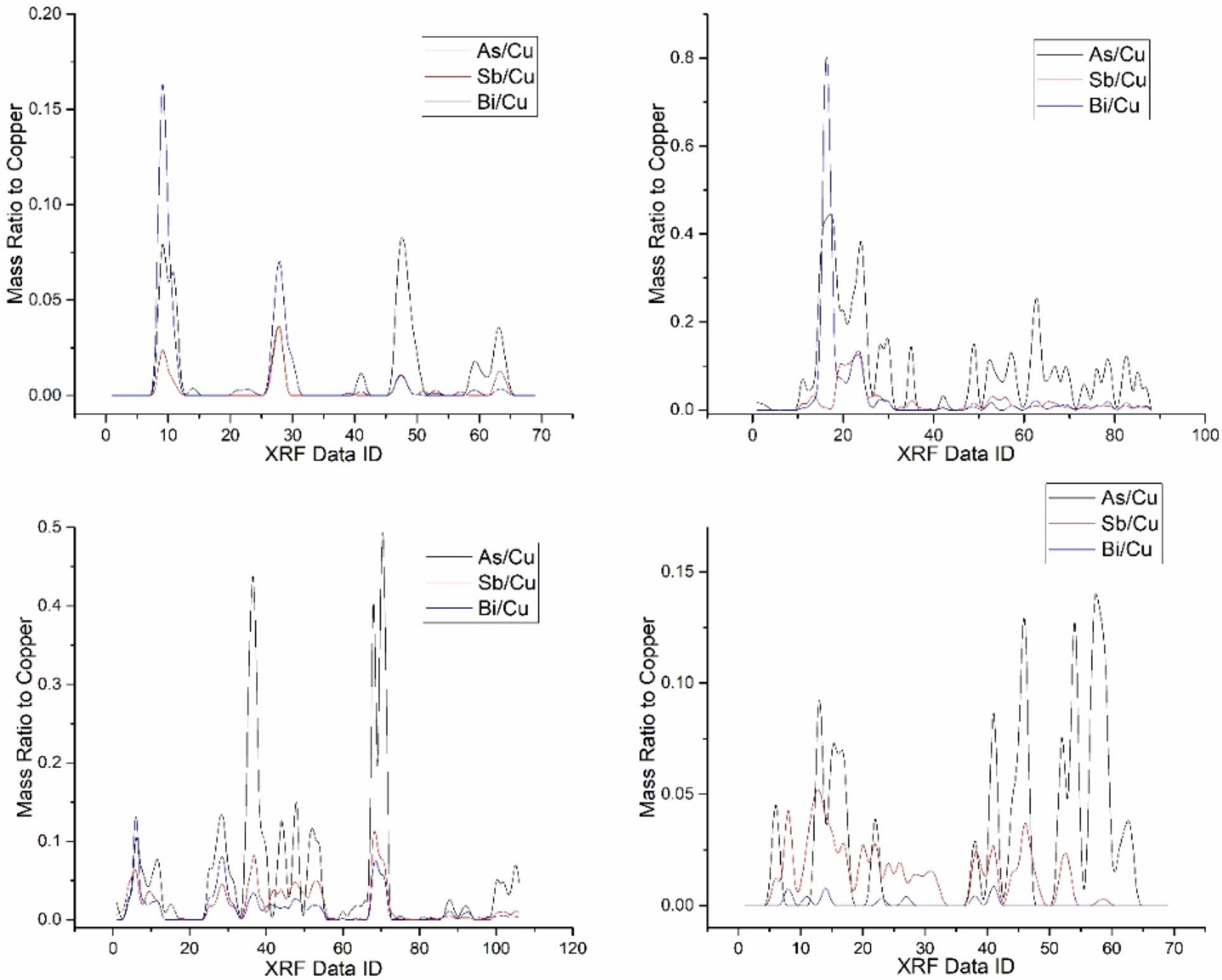
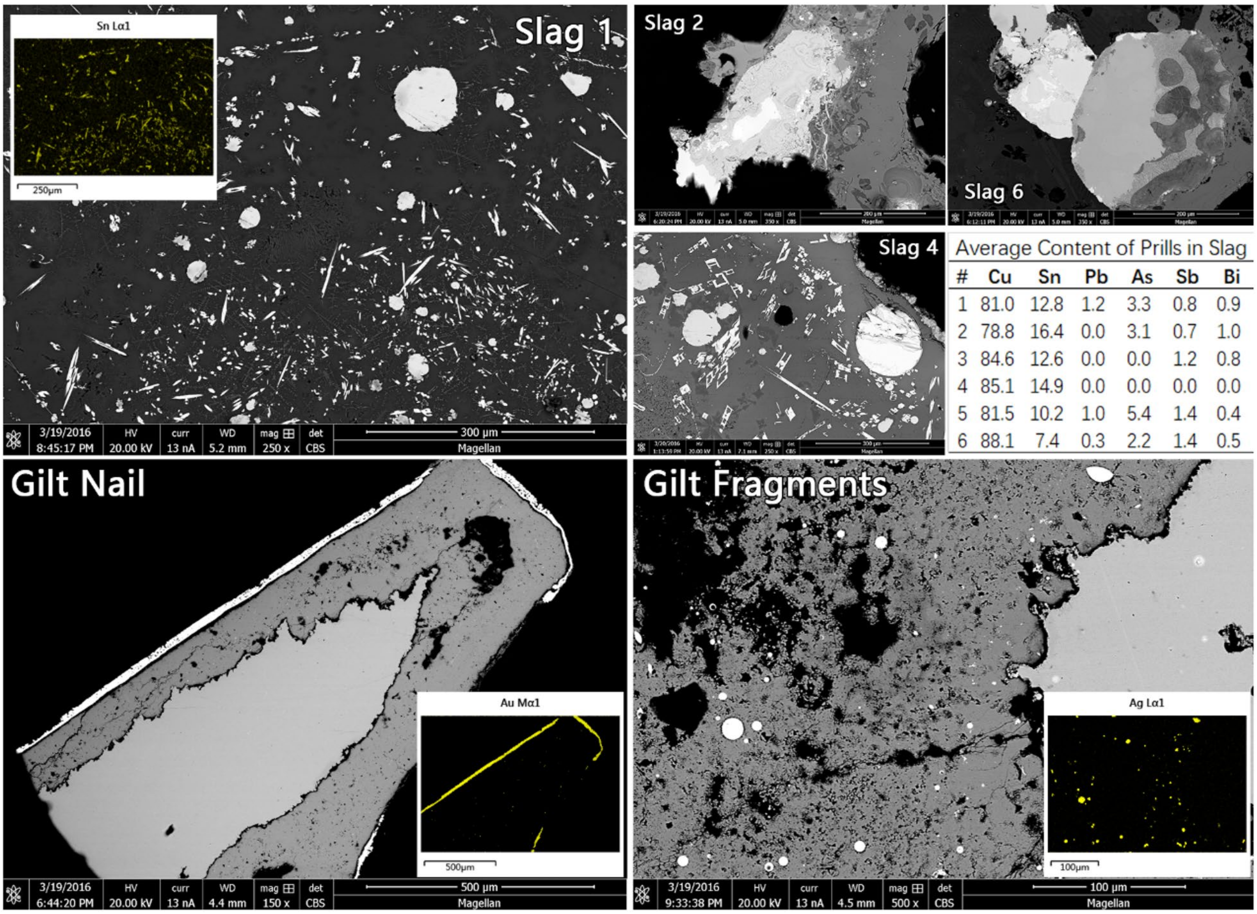


Fig. 8 The alloy elements content of Bohai artefacts. The data from the copper and bronze artefacts of four sites: SJ (UL), LTS (UR), Yanbian Museum (LL), Liudingshan Cemetery (LR)

is a crucial advantage of metal, especially for relatively high-value metals like copper, gold, and silver. However, this could sabotage the provenance studies via isotope and constitutional characteristics of the artefact to locate the specific resources. The mining and smelting sites and processing relics could not help us reconstruct the details of this 'chaine operatoire' [27] of the bronze artefact. We could still make an approximate approach to this indigenous bronze production tradition. Though the isotope and trace elements investigations were not involved in this research stage, the 'fingerprint' [29] of the element composition could still link the poly-metallic ore to the bronze casting. By considering the mixture/recycling [11] in the proceeding of the production, the content fluctuations on the paragenetic elements of copper ore like arsenic, antimony and bismuth of different batches of artefacts could not be just caused by the discrepancy between the different mineral vein deposit [30] as the raw material sources.

The widely unearthed cast bronze artefacts were heavy and simple, which reflected that the Bohai artisans had established a sophisticated bronze craft production system base on the vast stacks of the smelting copper slag and large-scale mining ruin. So the elite class might need imported high-class Tang-style art wares and gold to manifest the identity of a particular hierarchy. Most delicate copper and bronze ornaments were coating a 1–3 micrometres gold layer unearthed from the royal cemetery and capital city. Since the utilities of copper to produce implements and weapons were replaced by iron in the iron age, the social value of copper had been relocated by its aesthetic and currency properties; the flow of the copper materials should be discussed to comprehend the purpose of copper production and the



Average Content of Prills in Slag

#	Cu	Sn	Pb	As	Sb	Bi
1	81.0	12.8	1.2	3.3	0.8	0.9
2	78.8	16.4	0.0	3.1	0.7	1.0
3	84.6	12.6	0.0	0.0	1.2	0.8
4	85.1	14.9	0.0	0.0	0.0	0.0
5	81.5	10.2	1.0	5.4	1.4	0.4
6	88.1	7.4	0.3	2.2	1.4	0.5

Fig. 9 The SEM-EDS figure and data slag and artefact at upper capital city Ruin

quantity gap between the mining/smelting activities and the bronze artefacts. In that case, the scale of the mining and smelting of copper could be overcapacity for bronze artefact production only. According to the current archaeological discoveries, no mintage sites and other currency exchange evidence inside the Bohai kingdom, except for several bronze coins of the Tang Dynasty unearthed. Because the Central Plains society has continuously suffered from the copper shortage since the copper standard currency system was founded [31]. And the Bohai Kingdom maintained a relatively more stable relationship with Tang Dynasty than the Koguryo Kingdom in this region. It established a 'flourishing kingdom at eastern sea' approbated by the Central Plains people [32]. There was no conflict between Bohai and Tang recorded from the third king Mum ascended the throne (c.a 737 A.C), and LDG was very close to the Western Capital region of the Kingdom, which could provide both land and water routes to the Tang Dynasty [33]. All these situations above have provided commercial and geographical advantages for developing long-term and high-investment production like mining and smelting metals for exchange. So, the copper raw material trading could be the primary driver of the mining and smelting activities for the high-grade matte or blister copper ingot.

Conclusion

This research unveiled technical details about the bronze and copper artefacts unearthed from cemeteries and residential sites of the Bohai Kingdom. We had tentatively proposed the production chain of copper by combining the results of the slag, ore, and other relics from mining, smelting, and other production proceedings. The delicate and flimsy copper artefacts were considered to be imported and unearthed mainly from the capital region and royal graveyard, which could be highly related to the symbolism of dominance hierarchy. And the bronze artefacts shared similar alloy constitution patterns with the ore and slag from mining, smelting, and workshop sites were produced indigenously from the polymetallic copper vein within the Bohai realm.

Synthesised with the archaeological evidence and historical literature, we believed the driving force and the material flow of the mining and smelting of copper should be related to the shortage of mintage material in the Tang Dynasty. The blister copper/matte exchange maintains the proven mass-production scale and sophisticated technology of the bronze industry in the Bohai Kingdom. The prosperity of indigenous production of copper not only enhanced the metallurgy technology of the Bohai craftsman but also stabilised the geopolitical relations between the Bohai Kingdom and the Central Plains.

Abbreviations

LTS: Longtoushan Tombs; SJ: The Shangjing Cheng site; LDG: the Baoshan-Liudaogou site group at Linjiang City; GC: the Gaocheng site at Wangqing County; LH: Longhai; SG: Shiguo; GGZ: Gan Gouzi; GFG: Gan Fanguo; SEM–EDS: scanning electron microscopy coupled with energy X-ray dispersive spectroscopy; XRD: X-ray powder diffraction spectroscopy; p-XRF: Portable X-ray fluorescence spectroscopy.

Supplementary Information

The online version contains supplementary material available at https://doi.org/10.1186/s40494-022-00775-6.

Additional file 1. The p-XRF DATA of the copper and bronze artefacts of Bohai Kingdom.

Acknowledgements

Prof. FENG Enxue and prof. WANG Peixin of Jilin University, prof. FU Jiaxin of Northeast Normal University (CHINA), Ms Jia Ying and Mr AN Wenrong of the Jilin Province Department of Culture and Tourism have provided generous help for this research.

Author contributions

LQ is the principal discoverer of the site. YS and YJ is the archaeology and excavation counsellor of this study. SW has offered LTS excavation records. YL are supervisors of CL. CL is the major contributor to writing the manuscript. All authors read and approved the final manuscript.

Funding

This research was funded by the National Key R&D Program of China with the Grant number 2020YFC1521606.

Availability of data and materials

The datasets used and/or analysed during the current study are available from the corresponding author on reasonable request.

Declarations

Competing interests

The authors declare that they have no competing interests.

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Received: 21 April 2022 Accepted: 17 August 2022
Published online: 21 September 2022

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