ANALYSIS OF KUSHAN COINS (1–3 CENTURY CE) BY MULTI-SPECTROSCOPIC TECHNIQUES **

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Seven Indian copper coins of the Kushan Period (1–3rd century C.E.) were studied by multi-analytical techniques like WD-XRF, XRD, and FESEM-EDX to understand coin composition and the metallurgical process of fabrication. Analytical results reveal a compositional gradient in the distribution of elements between the external surface and the inner core of the coins, and the coins show isolated patches of surface mineralization leading to the formation of copper oxides, magnesium silicate, and silica. The analysis also showed the probable use of sulfide ore of copper for smelting and poor workmanship for this hoard. The impurities like sulfur, iron, silica, etc. could not be removed due to low firing temperature and improper poling during smelting. Striations have formed in the coin interior, with grain boundaries showing micro-cracks due to stress like punching or hammering with the die. The coins under study are a single-phase copper with associated impurities of Si, Al, P, and Mg in all coins and black spots of copper sulfide in coin inner core.

Keywords: *Kushan, striation, WD-XRF, copper sulfide, composition gradient, magnesium silicate.*

АНАЛИЗ КУШАНСКИХ МОНЕТ (1–3 в. н.э.) СПЕКТРОСКОПИЧЕСКИМИ МЕТОДАМИ

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Семь индийских медных монет кушанского периода (1–3 в. н.э.) изучены с помощью мультианалитических методов. Для установления состава монет и понимания металлургического процесса изготовления использованы волнодисперсионная рентгеновская флуоресценция, рентгеновская дифракция и полевая эмиссионная сканирующая электронная микроскопия с энергодисперсионной рентгеновской спектроскопией. Аналитические результаты показывают наличие градиента состава в распределении химических элементов между внешней поверхностью и внутренним ядром монет, а на поверхности монет обнаруживаются изолированные участки минерализации, приведшей к образованию оксидов меди, силиката магния и кремнезема. Анализ также показывает вероятное использование медносульфидной руды для плавки и плохое качество изготовления найденных монет. Примеси, такие как сера, железо, кремнезем и т. д., не могли быть удалены из-за низкой температуры обжига и плохого процесса монодоменизации во время плавки. Из-за напряжения в результате штамповки или удара молотком внутри монеты образовались бороздки с зернистыми границами и микротрещинами. Исследуемые монеты представляют собой однофазную медь с сопутствующими примесями Si, Al, P и Mg и черными пятнами сульфида меди во внутренних ядрах монет.

Ключевые слова: *Кушан, бороздчатость, волнодисперсионная рентгеновская флуоресценция, сульфид меди, градиент состава, силикат магния.*

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Introduction. Archaeometallurgical studies of ancient coins require proper understanding of metal physics as well as the analytical techniques employed. Based on analytical investigations achieved through WD-XRF and SEM-EDX, this paper highlights the existence of a compositional gradient in copper-based Kushan (1–3 century CE) coins. In ancient time the minerals chalcopyrite $CuFeS_2$ or bornite Cu_5FeS_4 were commonly used in the smelting of copper [1]. The chlorides and sulfur if present in the coin may cause active mineralization of copper owing to their property of corrosion accelerator [2, 3]. Many of [4] have reported that a concentration gradient is apparently uncommon in copper and brass coins. However, leaded bronzes often have concentration gradient due to segregation of lead-rich or tin-rich phases during coin solidification. The difference in chemical composition of the surface of the ancient coins from that of the core have now been well documented [4, 5]. The radiation techniques like XRF, EPMA, and PIXE commonly applied for the analysis of old coins give the surface composition of limited depth (from few micrometers to a few tens of micrometers) [6, 7]. However, if the composition of the surface layer of an object is altered, the radiation techniques may provide inaccurate data from the surface of the objects that is not at all representative of the bulk composition. The main reason of surface inhomogeneity in metals is the depletion of a more chemically active phase in contact with the less active phase [8], active corrosion, or surface enrichment of the objects [9, 10]. The inhomogeneity in coins may also result from remelting of old coins, diluting precious metal with the base metal in antiquity, certain impurities in the ores that are difficult to remove, or alloy addition made intentionally to lower the melting temperature, etc. Due to the aforesaid reasons, several incomplete and sometimes unreliable analysis have been reported for copper-based coins of the Roman Republic [11]. However, it is stated that removal of a thin layer of the metal at the surface of the coin usually expose the surface, which is representative of the composition of the entire coin [12], but there are many exceptions [13].

Although the technical and numismatic complexity of Kushan coins is well documented [14], there is a lack of data on the chemical composition and copper percentage due to the small number of investigations [15]. In the literature, there is no document about the chemical composition or metallurgical processes applied for the fabrication of these coins. The main aim of this work is to evaluate the composition of Kushan copper coins (1–3 century CE) by using highly sensitive and reliable WD-XRF technique in preference to ED-XRF most commonly used for this purpose. With WD-XRF, it was attempted to perform analysis on areas that were not affected by the corrosion layer, and analysis was carried on areas as large as possible in order to avoid any surface inhomogeneity from the bulk. In the present work, it was also attempted to evaluate the core composition on cross-section of the specific fragments of three Kushan coins at a different accelerated voltage of incoming electrons. The performed SEM-EDX analysis was compared with the one observed through WD-XRF with the purpose to differentiate the core composition from the exterior coin surface. The main advantage of the scanning electron microscope coupled with microanalysis is to visualize the interior of the coins and its composition. In addition to elemental composition and their distribution on the surface as well as cross-sectional analysis of the coins, we also studied the corrosion features and surface behavior by X-ray diffraction (XRD) to explore natural patina on these coins. This has helped to diagnose the material degradation of Kushan coins. This paper highlights the compositional gradient of Kushan coins using multi-analytical spectroscopic techniques to understand the metallurgical process and coin composition.

The tribal confederacy known as Yuch-Chih in Chinese sources extended the kingdom in 1–3 Century CE and founded the Kushan kingdom [16]. Kushans minted a large number of coins, suggesting a well-developed money economy. They mainly struck gold and copper coins and silver coins were only minted in the lower Indus area due to acute scarcity of silver in other parts of the empire [17]. A noteworthy feature of the currency system of the Kushans is that their gold and copper coins were meant for circulation throughout the empire. Unlike the Indo-Greeks, Scytho-parthains, early Kushans and others, the Kushan coins were in circulation outside their empire and as per literature [18], the Kushans were responsible for issuing the first imperial coinage of India.

Experimental. The copper coinage of the Kushan dynasty under study was probably struck in northwest India and in the author's (MS) private collection. On the obverse of most coins (Fig. 1), crowned diademed standing king, holding standard and sacrificing at altar at left, is seen. On the reverse site in some coins, two-armed standing Shiva holding trident and bull Nandi left behind is seen. In some coins on the reverse side, goddess of plenty Ardochsho enthroned holding diadem and cornucopia is seen (Fig. 1). The diameter of the coins varied between 17.96 to 21.78 mm and thickness between 3.02 to 3.74 mm. The weight of the coins varied from 6.3 to 8.3 g and clippers were used to cut the coins to the most approximate weight around 7.0 g.

Fig. 1. Obverse and reverse view of the Kushan coins.

The X-ray fluorescence (XRF) provides one of the simplest, most accurate, and widely used analytical technique for determination of elemental composition on a wide variety of materials. In the wavelength dispersive XRF (WD-XRF) the fluorescence X-rays emitted by the sample is further dispersed by Bragg's diffraction from an analyzer crystal. The resolution, in this case, is not determined by the detector being used. The much higher spectral resolution afforded by WD-XRF implies that there are very little spectral overlap and lower background intensity. The much lower background in WD-XRF is clearly an advantage in detecting very low signals from a low concentration of sample elements. Hence, as compared to ED-XRF, the WD-XRF not only allows obtaining much more accurate relative concentrations from a multi-element sample but also allows detecting elements present at much lower elemental composition. While almost all previous analysis of early Indian coins depended on the ED-XRF technique [15], we provide a more comprehensive and accurate evaluation of Kushan copper coins using a combination of WD-XRF, relating anode X-ray diffraction (XRD) and energy dispersive X-ray analysis (EDX) based on field-emission scanning electron microscope (FE-SEM). The seven Kushan coins under study were analyzed by a Rigaku supermini-200 high power, sequential wavelength WD-XRF spectrometer for high-resolution elemental analysis in the atomic number range of oxygen (^{8}O) through uranium (^{92}U). The supermini-200 WD-XRF system has a high power (200 W, 50 kV) Pd X-ray tube, resulting in lower detection limits and shorter measurement times. Depending on the wavelength range it uses the following three analyzing crystals: Li F 200 (for Ti-U) and PET (for Al-Ti), and Rx25 (for F-Mg). Similarly, it uses two different X-ray detectors, a gas flow proportional counter for light elements, and a scintillation counter for heavy elements. XRD studies of all the coins were carried using Rigaku Smartlab X-ray diffractometer with 9 kW rotating anode X-ray generator. For XRD spectra the surface of the sample showing bluish coloration was targeted. Although XRD spectra were taken for all the coins in the present study, we have shown spectra of four coins only (Nos. 1, 2, 3, and 7). For SEM-EDX of the coins, a Zeiss ultra-Field emission scanning electron microscope (FESEM) with an Oxford EDX analyzer was used to study the cross-sectional microstructure. As the Kushan coins were drawn from the private collection of the author (MS), three coins (Nos. 2, 5, and 6) were allowed for sectionning to investigate the microstructure of the coin interior.

Result and discussion. *Analysis by WD-XRF.* The copper coin under study is a single-phase copper alloy used in the manufacture of the coins. The major elements identified in these coins are Cu, Mg, and Si, whereas P, S, Cr, Mn, Ti, Ag, and Ba are present in trace quantities in some coins only. The chemical composition of the coins derived from sensitive WD-XRF is shown in Table 1. At least 10 measurements both on the obverse and reverse sides of the coins (five on each side) were taken and the values averaged in this study. As the coins were not much corroded, the WD-XRF was targeted on non-corroded surfaces on a larger surface area. The copper percentage on the obverse side varied between 93.70 to 97.20% (Table 1). On the reverse side, the percentage of copper varied between 92.70 to 97.10%, showing copper as the main constituents of the coins. Except for coin No. 4, none of the coins showed the presence of any sulfur on the exterior surfaces. Coin No. 4 shows variations of sulfur from 1.52% on the obverse to 6.39% on the reverse side. This is interesting because, through cross-sectional analysis of coins (Nos. 2, 5, and 6) by SEM-EDX, we

have observed the presence of sulfur in the interior of the coins with the sulfur percentage at some spots as high as 14.63 to 25.35%. The sulfur as corrosion accelerator might have been lost from the external surfaces of most coins due to its high volatility and segregation of sulfur grains on the isolated spots that were lost during handling in the circulation of coins. None of the coins showed any traces of iron on the external surfaces wherein iron was noticed through cross-sectional analysis of coin No. 2 from traces to as high as 8.15%. All the copper coins are free from the presence of any chloride, K, Ca, and iron on the external surfaces. The elements Cr (No. 2), Ti (Nos. 3, 5, 6, and 7), and Mn (Nos. 3 and 5) are present in traces on the surface. Silver is present in traces (0.12%) on the obverse side of coin No. 6, which may be impurity/contamination during smelting. The obverse/ reverse composition of all the coins show the presence of silica in minor quantities from 0.49 to 3.03%. Probably, silica could not be removed during the metallurgical process, which points towards poor workmanship for this hoard. Phosphorus in traces (0.11 to 0.14%) is present in all the studied coins. The presence of silica and alumina (0.18 to 0.94%) in all the coins and sulfur in coin No. 4, revealed through WD-XRF analysis, indicates poor workmanship of coins as inclusion like sulfur, silica etc. could not be removed during smelting. Barium has been detected in coin No. 7 only. Elements like Fe, Ni, In, Pb, Bi, As, Ca, and chlorides were not detected in any of the coins for this hoard as reported [19] for other hoard Kushan coin. In most of the Kushan coins, copper was rarely used as pure metal and preferably mixed with Zn or Sn to reduce the smelting temperature and save in cast. Interestingly, copper was used as pure metal for this hoard as the presence of Zn or Sn were not detected in any of the studied coins.

TABLE 1. Elemental Analysis (mas.%) by WD-XRF of Kushan Coins From Obverse and Reverse View

Element		Cu	Si	Al	Mg	P	Ti	S	Cr	Ag	Mn	Ba
Coin 1	Obverse	97.2	1.90	0.72	0.13	0.09						
	Reverse	97.1	1.69	0.68	0.48	0.08						
Coin 2	Obverse	97.1	2.12	0.52	–	0.12						
	Reverse	97.0	2.05	0.66	0.25	–	–	–	0.032			
Coin 3	Obverse	95.4	3.03	0.94	0.24	0.13	0.05	–	–	–	0.02	0.14
	Reverse	96.6	2.19	0.74	0.32	0.11	–	–	–	–	–	–
Coin 4	Obverse	96.9	1.08	0.4	–	0.12	–	1.52	–			
	Reverse	92.7	0.49	0.18	0.18	0.09	–	6.39	–			
Coin 5	Obverse	95.3	3.01	1.06	0.44	0.14	0.04	–	–	–	–	
	Reverse	95.4	3.00	0.89	0.54	0.14	–	–	–	–	0.02	
Coin 6	Obverse	93.7	3.46	1.02	1.45	0.15	0.06	–	–	0.12		
	Reverse	95.4	2.59	0.93	0.94	0.09	–	–	–	–		
Coin 7	Obverse	95.6	2.88	0.77	0.36	0.11	0.06	–	–	–	–	–
	Reverse	97.0	2.11	0.54	0.2	0.1	–	–	0.02	–	–	0.17

From the comparative study of the composition of Kushan coins from the eastern part of India analyzed through PIXE technique [14], it is observed that the studied coins do not contain iron, nickel, zinc, lead, and bismuth on either surface as minor or trace elements, showing better metallurgical process or selection of copper ores for smelting. The presence of lead from 0.56 to 13.39% detected in Eastern India Kushan coins of Orissa led to the brittleness of the coins. We have not observed the presence of lead in our studied coins either on the surface or in coin core, showing better refinement or selection of ores of this hoard. Based on physical features, the numismatists have expressed the north-west Indian origin of these coins probably from mint located somewhere in Kashmir state. Poor quality control must have been exercised during the metallurgical process of Kushan coins as the coin varied in chemical composition and weight leading to the remnant of sulfur and other impurities within the coin core and thus compositional gradient.

X-ray diffraction analysis. All the coins were studied under X-ray diffraction to characterize the corrosion features, surface behavior, and the nature of patina on these coins. As all the coins showed similar XRD peaks in this study, only the XRD pattern of four coins Nos. 1, 2, 3, and 7, are presented in Fig. 2. From the XRD data, it is observed that cuprous oxide (Cu_2O) and cupric oxide (CuO) have invariably formed on all the coins along with magnesium silicate (Nos. 1, 2, and 3) and silica (No. 7). More than 95% of copper and the rest of impurities allowed strength and resistance to corrosion for the coins. The high background of XRD images indicates the presence of corrosion product in all the coins. Visual appearance also gives minor bluish coloration on the surfaces at isolated spots on the coins. Also, the impurities of sulfur played an im-

portant role in the deterioration of the surfaces. Cuprous oxides Cu_2O was detected as a major corrosion compound along with magnesium silicate. Cuprite, the first and most widely occurring alteration mineral of ancient copper, is formed during burial or bad storage due to exposure to oxygen or moist air. Cuprite is known to play a decisive role in the protection of corrosion layers on copper, but the increase in oxygen pressure breaks its conformity and provokes dissolution. For the Kushan coins, it appears that the copper oxide matrix has provided enough surface protection, which saved the coin as we did not notice any bulk corrosion product on either surface of the coins. In the XRD spectra, silica peaks are also seen, which seems to have merged with the magnesium silicate peaks, resulting in widening of the peaks. The other peaks of copper oxide are seen to be very close to magnesium silicate peak in the XRD spectra.

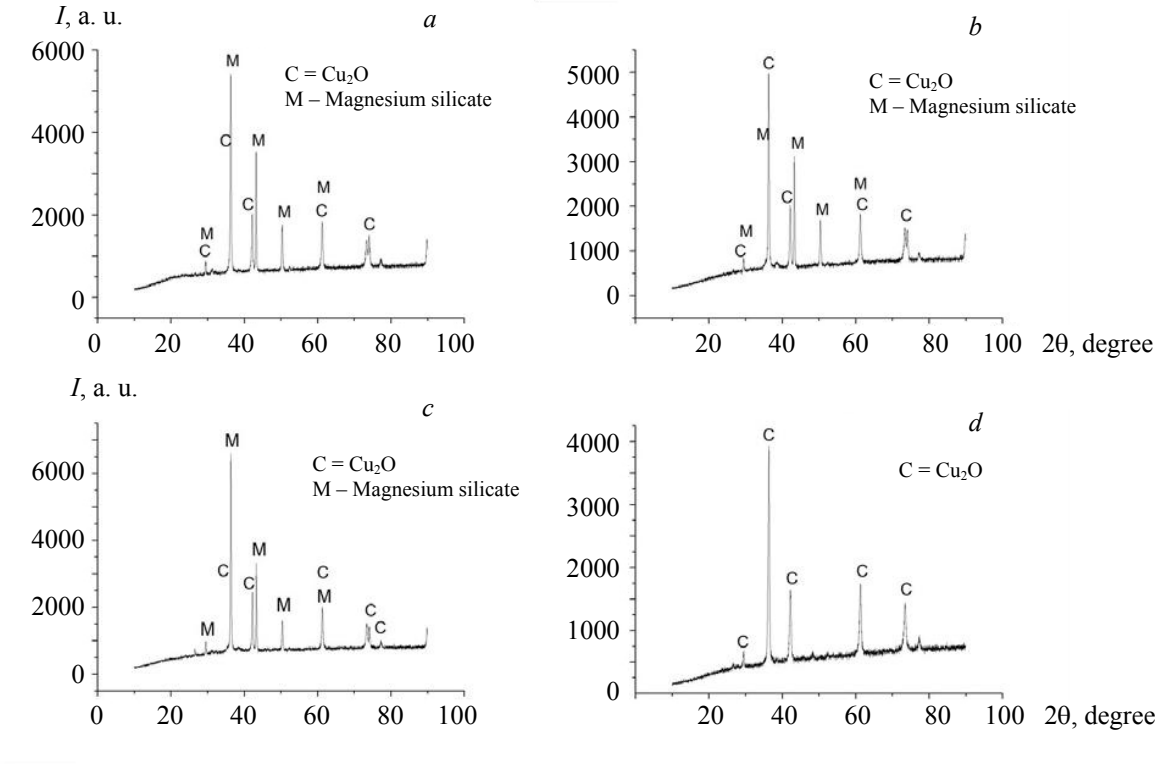
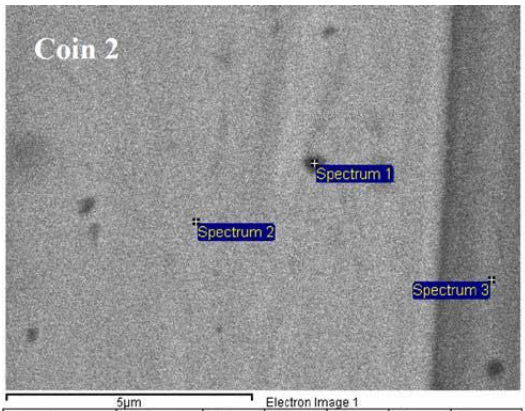


Fig. 2. XRD analysis of the Kushan coins No. 1 (a), 2 (b), 3 (c), and 7 (d).

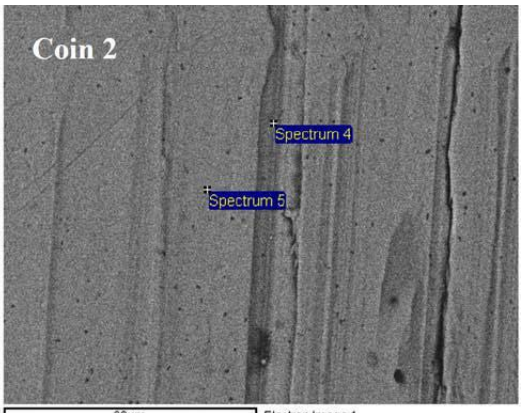
SEM-EDX analysis. A combination of instrumental techniques provides far better information about the composition of the coins that can answer many archaeological queries besides indicating the types of ores exploited for the minting of coins. Besides, the knowledge of original alloy composition can also reveal the fineness of the coins and discriminate between the original and a fake one. Therefore, high magnification field emission scanning electron microscope coupled with EDX was applied for the detection and quantitative estimation of areas of the coins that have not been affected by any corrosion to widen the scope of this investigation. The Kushan coins Nos. 2, 5, and 6 were allowed for sectioning for analysis by SEM-EDX. The results of SEM-EDX photomicrograph along with quantitative EDX data recorded from different spots of coin core is presented in Fig. 3. From the photomicrograph, it is clear that the Kushan coins are made of a single phase of copper, and no other metal phase co-exist in it. The quantity of Sulfur varied from 0.8 to 8.15% in coin 2 (Fig. 3a), 0.59 to 25.35% in coin No. 5 (Fig. 3b), and 0.6 to 8.6% in coin No. 6 (Fig. 3c) when the beam was targeted to analyze black inclusions in the copper matrix. One black inclusion (No. 5) showed the Cu to sulfur ratio as 3:1, indicating the fractured inner surface may contain copper sulfide. Some microcracks at high magnification along with black spots (sulfide inclusions) on fractured surface co-relates and confirm the finding by XRD. The microcracks in the coins may be due to stress (punching or hammering with die). It has been reported that a sulfur content up to 0.33% is due to buried soil conditions, whereas a sulfur content greater than 15% may be due to a black porous area in which there is a large content of Cu with sulfur [20, 21], which denotes a mixture of copper sulfide. The presence of other inclusions of silica and alumina in coin Nos. 2, 5, and 6 (blackish spots) and from traces to very high percentage of iron (No. 2) in-

dicate poor workmanship in the fabrication of the coins as inclusion like sulfur, silica, iron, etc. could not be removed during smelting. Probably poling during smelting could not be done properly, resulting in a remnant of sulfur within the bulk. The presence of impurities like sulfur, iron, silica, etc. in the copper matrix also points towards the use of copper-iron-sulfide like chalcopyrite $CuFeS_2$ or bornite Cu_5FeS_4 for extraction of copper as sulfide ores of copper were of importance in ancient times for the smelting of copper [1].

a

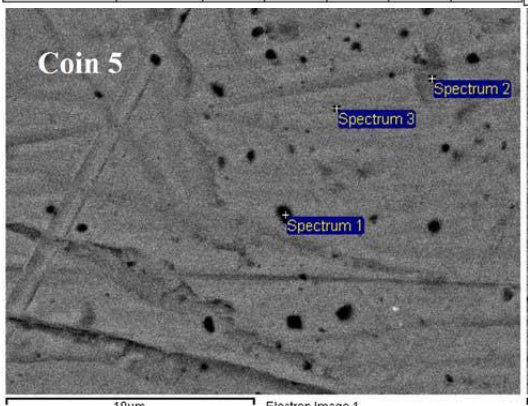


Spectrum	In stats.	Al	Si	S	Fe	Cu
Spectrum 1	Yes	2.00	1.44	2.25	8.15	86.15
Spectrum 2	Yes			0.76		99.24
Spectrum 3	Yes			2.56		97.44

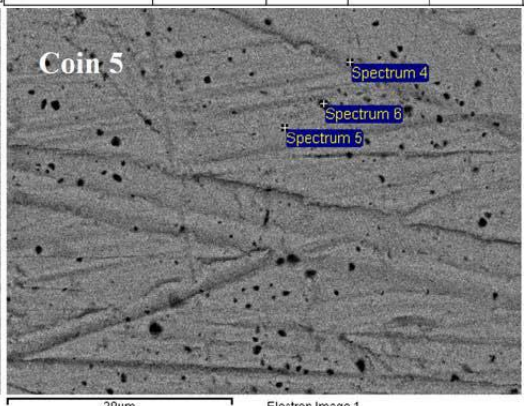


Spectrum	In stats.	Al	S	Cu
Spectrum 4	Yes	2.81	1.99	95.19
Spectrum 5	Yes		0.80	99.20

b

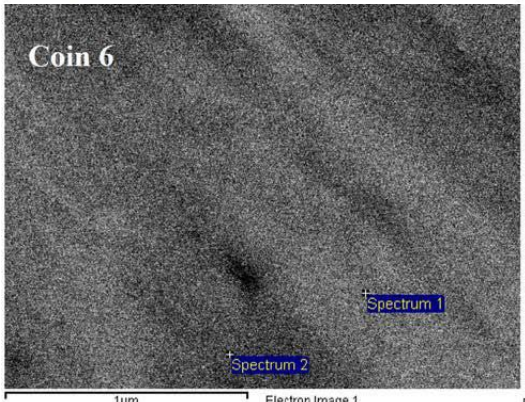


Spectrum	In stats.	Si	S	Cu
Spectrum 1	Yes	39.30		60.70
Spectrum 2	Yes		25.35	74.65
Spectrum 3	Yes		1.17	98.83

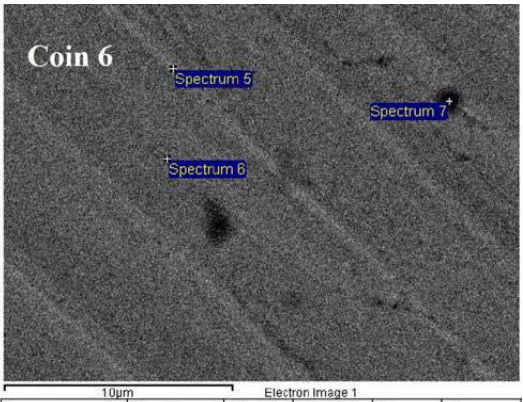


Spectrum	In stats.	Si	S	Cu
Spectrum 4	Yes	0.91		99.09
Spectrum 5	Yes			100.00
Spectrum 6	Yes	24.54	0.54	74.91

c



Spectrum	In stats.	S	Cu
Spectrum 1	Yes	4.03	95.97
Spectrum 2	Yes	8.59	91.41



Spectrum	In stats.	Al	Si	S	Cu
Spectrum 5	Yes			1.20	98.80
Spectrum 6	Yes			1.44	98.56
Spectrum 7	Yes	7.25	35.06		57.70

Fig. 3. SEM-EDX photomicrograph of Kushan coins Nos. 2 (a), 5 (b), and 6 (c).

Conclusion. The multi-analytical investigations of Kushan copper coins confirmed the probable smelting of sulfide ores of copper for extraction of metal. Due to poor workmanship and poor quality control, impurities like silica, iron, and sulfur could not be removed from the bulk of the composition during smelting. The existence of compositional gradient was observed in the coins as WD-XRF did not show the presence of sulfur and iron on the coin surfaces. The coins show limited corrosion at isolated spots and were in a fairly good state. Pure copper metal instead of bronze or brass was used for this hoard in the fabrication of coins. Magnesium is present only on either surface of the coins and not in the inner core. Magnesium has mineralized to magnesium silicate on the coin surface (XRD result). The present study shows improper smelting of ores for fabricating the Kushan coins, as evidenced by the presence of sulfur from traces to about 25.35% in coin core. A very high percentage of silica (from 1.44 to 39.30%) was detected on selected spots in the coin core due to improper poling and poor refining process. The studied coins characteristically differ from the other reported Indian Kushan coins in composition and fabrication.

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