THE BLACK SURFACE COATING OF ANCIENT POTTERY 
EXCAVATED FROM WARI-BATESHWAR AND 
MAHASTHAN, BANGLADESH

Dilruba Sharmin*, Takayuki Honda** and Fumio Okada***

Quotation: Black coloured ancient potsherds from Bangladesh were analysed to investigate the black coating technique with observing thin sections by optical microscope, scanning electron microscope and pyrolysis-gas chromatography/mass spectrometric (Py-GC/MS) analysis with ATR spectroscopy. Single-staged, two-staged and multi-staged surface coatings were identified in the analysed black coloured potsherds by thin section observation. The Py-GC/MS method identified oil and lacquer on the single-staged coating, oil on the top coating of NBPW and solely oil on the multi-staged coating of the analysed potsherds. ATR mapping determined the application technique of the surface coating.

Key words: Northern Black Polished Ware, Bangladesh, Thin section, Slip, Lacquer, Drying oil, Py-GC/MS, ATR spectroscopy.

Introduction
Identifying the characteristics of the black coating found on black coloured pottery excavated from Bangladesh is the primary focus of the present study. Several types of pottery have been excavated from many archaeological sites in Bangladesh. These pottery goes by various names, including ‘Northern Black Polished Ware’ (NBPW), ‘Black Slipped Ware’ (BSW), ‘Black-and-Red Ware’, ‘Rouletted Ware’, ‘Glazed Ware’, and ‘Knobbed Ware’.1 Until now, however, no diagnostic analysis regarding surface coating technology has been carried out in Bangladesh.

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In the present study scientific analyses were conducted on the surface coating of black coloured ancient pottery, which will enable us to evaluate the technological advancements of the past. Initially, potsherds of the black coloured pottery were collected from two early historic sites of Bangladesh, and surfaces of those potsherds were observed using a binocular microscope and a scanning electron microscope. Next, thin sections were prepared from collected potsherds for microscopic observation. Single-staged, two-staged, and multi-staged surface coatings were identified by the thin section observation. Scientific analysis of this cultural material is important not only to distinguish the technology or characteristics, but also to understand the spread of regional culture related to the analysed material. For that purpose, the collected black coloured potsherds were analysed by Py-GC/MS and ATR spectroscopy. The results show that some of the potsherds contained an organic substance like oil on their top coats; two pieces of potsherds contained solely oil; and one piece of analysed potsherd contained oil with a small amount of lacquer in its surface coating. ‘Oil coated’ or ‘lacquer coated’ pottery is not familiar in the ancient pottery of Bangladesh. Therefore, the results of this primary study could provide new information for assessing the technological development of the ancient pottery of Bangladesh as well as south Asia.

SITES, RESEARCH MATERIALS AND PREVIOUS STUDIES

Sites
Potsherds were collected from the excavated sites, namely, Wari-Bateshwar, in the Narshingdi district and Mahasthangarh, in the Bogra district [Fig. 1]. Narshingdi is located in central Bangladesh, 50 km northeast of Dhaka, and Bogra is situated in Northern Bangladesh. Wari and Bateshwar are two adjacent villages in the Narshingdi district. The soil of the entire area is red and contains pebbles. Wari and Bateshwar stand on relatively flat ground, which is considerably higher than the surrounding area. There are marshlands between the two villages to their north, northeast, east and southeast. The confluence of the Arial-Kha and Brahmaputra rivers is 4 km northeast of Wari. The shrunken and nearly dry channel of the river Koira borders Wari in the north. Sitalakhya, another major river in the vicinity, flows some 35 km west of Wari and Bateshwar.²

Fig. 1. Map showing the location of Bangladesh in South Asia, the Mahasthangarh and Wari-Bateshwar archaeological sites in Bangladesh and collected potsherds.

The archaeological importance of this site was first brought to light by a local school teacher named Md. Hanif Pathan in 1933. Later, his son, Md. Habibullah Pathan, an amateur archaeologist, took initiative to collect the antiquities and study them. A small-scale trial excavation of this site began in 2000 (February 4th - April 24th), carried out by the International Centre for Study of Bengal Art (ICSBA). This excavation discovered significant artefacts, including NBPW. These discoveries place the active period of Wari-Bateshwar in the early historic period. Excavation work at the Wari-Bateshwar site is still being carried out under the supervision of archaeologist Dr. Sufi Mostafizur Rahman. Various types of artefacts are being discovered at every phase.

The Mahasthangarh site is located in Mahasthan village in the Bogra district in northern Bangladesh. During early history (c. 500 BCE to 550 A.D) and the early medieval period (c. 550 A.D to 1200 or 1300 A.D), the Bogra district was a part of the kingdom of Pundravardhana and was part of the Gauda Empire. The early historic and early medieval sites of this district are situated on the red-bed Barind tracts, which are slightly elevated terraces on the alluvium. This area is higher in elevation than the surrounding plains and forms a distinct (and relatively flood-free) physiographic unit. The elevation of the district ranges from 15 to 25 m above mean sea level. The plains are dissected by erosion, the ridge tops between the depressions are almost level, and the valley sides and floors have been terraced to allow cultivation. The ridge tops, which preserve evidence of

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human occupation in the early historic and early medieval periods, have been designated as ‘sites’. Most of the ridge tops were occupied by human groups during the early historic and medieval periods. The majority of the sites consist of a mound containing burnt-brick structures representing a stupa, a temple, viharas, or other structures. The term ‘site’ has been used in a broad sense and includes the mounds that lie within it.

The size of the sites in the Mahasthan area has been calculated on the basis of the area over which the found artefacts are distributed. As most of the sites of the Mahasthan area are covered by modern occupied structures, the distribution area of artefacts and, therefore, the size of sites cannot be measured precisely. During exploration, 135 sites were found in an area of 2920 km² in the Bogra district, with Mahasthan as one of the sites. For the present study, potsherd samples were collected from Mahasthan. The Mahasthan citadel is situated in the village of Mahasthan and is surrounded by the river Karatoya on the east and a moat on the other three sides. This moat is locally known as garh. No archaeological remains prior to the late fourth century BCE have been recovered from the region of Mahatshangarh, which suggests that the initial population selected this area and quickly built the site, possibly as a trading centre, given its favourable location on the banks of the Karatoya river.

The archaeological site in the Mahasthan area was discovered by Sir Alexander Cunningham’s surveys in the late nineteenth century (1879, precisely). A framework for Mahasthan history was established in 1929 through a survey of written sources at a time when regular excavations were starting at the site under the direction of K.N. Dikshit (Archaeological Survey of India) in 1929-1930 and 1934-1936, with a sketch plan published by P.C. Sen. No final report on the ASI excavation was ever issued. In 1960-1961, official excavations were conducted by the Pakistan Department of Archaeology and Museums. Short soundings were

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also made in 1965 and 1966. Since 1988, the Directorate of Archaeology of Bangladesh has conducted several periods of excavation, conservation and restoration. Unfortunately, most of the results of over 50 years of research regarding Mahasthan remain unpublished. A joint French- Bangladeshi venture excavation was carried out from 1993-1999 in the Mahasthangarh area, and the first interim report was published in 2001.

Research Materials

A total of 30 pieces of black coloured potsherds were collected for this study. Among these, 18 were collected from the Wari-Bateshwar archaeological site. The other 12 pieces were collected from the Mahasthangarh site.

All the potsherds of this study were initially collected as ‘NBPW potsherds’, but observation suggested that 17 pieces of the Wari-Bateshwar potsherds were similar [Type A], and the remaining one piece was different from the others [Type B]. These potsherds were primarily examined by a binocular microscope (Olympus SZH10) with Fiber light FL-50. The observation suggested that 17 pieces of the potsherds had black glossy coating on both their inner and outer surfaces [Type A] [Fig. 2a & 2b].

The one remaining piece was not glossy and was badly marred by piled up clay on both sides [Type B] [Fig. 3a & 3b]. The existing surface coating of Type B was thick and fragile and detached from the body surface.

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7 Ibid.
8 Ibid.
Twelve pieces of black coloured potsherds were collected from the Mahasthangarh site. Among these, 10 pieces of potsherds possessed the same characteristics like Wari-Bateshwar Type A and tiled as Type C [Fig. 4a & 4b].

The remaining two pieces were different from the other 10. Binocular observations of these two pieces revealed they were different from each other [Type D and Type E]. In the potsherd of Type D, the binocular observation found that (a) the applied coating was intact, (b) it was not uniform over the entire surface and (c) it was not detached from the surface [Fig. 5a & 5b]. In the Type E potsherd, the black coating was found on a small portion of the inner side, and the binocular observation revealed that the coating was detached from the body surface [Fig. 5c & 5d].
Previous studies regarding NBPW

The initial recognition of what would become known as ‘NBPW’ was by Sir John Marshall (ASI-AR 1904-5) at Sarnath, in India (1904).9 Since then, NBPW has been discovered in various regions of Pakistan, India, Bangladesh, Nepal and Sri Lanka. The Northernmost site of NBPW is in Udegram in Pakistan; the southernmost site is in Anuradhapura in Sri Lanka; the westernmost site is in Prabhas-Pathan in India; and the easternmost site is Wari-Bateshwar in Bangladesh.10 In 1946, after the excavation of Ahichhatra, India, Wheeler and Krishna Deva (1945) first proposed the term ‘Northern Black Polished Ware’. They also discarded the view of J. Marshall, who regarded NBPW as a kind of ‘Greek Black Ware’ on the grounds that at Taxila, the NBPW was mainly of the

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pre-Greek period. Asok Datta (1999) mentioned that the area lying to the east at the confluence of the Ganga and Yamuna rivers, known as the middle Ganga Valley and where there is very fine alluvium soil, is possibly the birthplace of NBPW.

NBPW was potted on a fast wheel and is made of fine-grained clay with little tempering material. NBPW specimens have a highly glossy surface that can easily be distinguished by the fine quality of its manufacturing technique.

In the last 65 years, many researchers, including Sana Ullah (1946), B.B. Lal (1956), K.T.M. Hedge (1966, 1976, 1978), H.C. Bhardwaj (1973, 1979), Mitchell (1979), K.J.S. Gillies & D.S. Urch (1983) and Robert Harding (2004), have proposed hypotheses regarding the black glossy coating found on NBPW. Sana Ullah (1946) and K.T.M. Hedge proposed that the black colourations of NBPW are due to iron compounds present in the material, whereas B.B. Lal (1955-1956), H.C. Bhardwaj (1979) and Mitchell (1979) claimed that carbon is the dominant black colourant in NBPW.

An examination of NBPW (bichrome and monochrome groups) excavated from Rajgir, India, by Robert Harding (2004), found that the examined samples had two slips, with the upper surface being approximately one µm in thickness. The upper coating was observed to be thinner where an edge was exposed: it was also indicated by the presence of air bubbles on the sherd surface. Despite many hypotheses and scientific analyses, the production method and surface coating technology of NBPW has not been settled satisfactorily.

In Bangladesh, NBPW has been excavated from two early historic sites, Wari-Bateshwar and Mahasthangarh. NBPW from the Mahasthangarh site has been dated from c. the fourth century BCE to the first century A.D. The forms

of the NBPW found at Mahasthangarh include dishes, cups (with or without a lip), bowls and beakers. Most of the examples of NBPW excavated from the Mahasthangarh site are black, but there are also red, silver, and golden colours.\textsuperscript{17}

The time period for the Wari-Bateshwar site has been set at between \textit{c.} 700 BCE and 100 BCE/50 A.D \textit{for the sub-continental site}.\textsuperscript{18} A series of recent radiocarbon dating results from Ayodhya in Uttar Pradesh, India, suggest that the NBPW phase could go back to as early as \textit{c.} 1000 BCE.\textsuperscript{19} The examples of NBPW excavated from the Wari-Bateshwar site are dishes, bowls and spouted jars, all of which possess black and grey cores: their surfaces are commonly a lustrous black colour, however light chocolate, light silver and light red potsherds have also been found.\textsuperscript{20}

\textbf{METHODS OF ANALYSIS}

Scientific analysis of the surface coating is important for identifying the origin of the coating and the characteristics of the materials as well as for the future conservation. The analysis is carried out by four basic methods.

First, the surfaces of the collected potsherds were observed using a scanning electron microscope (SEM) (Hitachi Miniscope TM-1000).

Second, thin sections were prepared using the ‘Polished thin section method’ for observation in a camera-attached optical microscope (Nikon Digital Camera DXM 1200F, Japan) and scanning electron microscope (SEM). The ‘Polished Thin-Section Method’ is used in this study to prepare the thin section of the collected potsherds.\textsuperscript{21} This method is advantageous for the following reasons: (a) it requires little experience; (b) it is relatively inexpensive; (c) it requires little variation in technique for different types of material; (d) the prepared thin


\textsuperscript{19} U. Singh, \textit{A history of Ancient and Early Medieval India}, (India 2008), p. 260.


sections are clear for identifying the surface coating, including the ground coat and top coat, with the quality of the used clay; and (e) the prepared thin sections can be stored for many years at normal room temperature. Thin-sectional microscopic specimens can evaluate the making technique, the quality of the used clay, and the characteristics of the applied slip and coating.

The potsherds to be used for thin sections were divided into several pieces (approximately 2.5-4.0 mm in measurement). The potsherd samples were then embedded in epoxy resin. Next, the surfaces of the embedded samples were ground flat and polished with wet-type sandpaper. The upper surfaces were observed with a stereoscopic or metallurgical microscope, and the samples were attached to the slide glass with epoxy resin (Adeka resin). The other side of the sample was ground and polished until it was approximately 20 µm in thickness. Then, the prepared slide was observed using the optical microscope and SEM. All of the microscopic analyses were carried out in the laboratory of the Art Research Institute at the Kyoto University of Art and Design in Kyoto, Japan.

Third, pyrolysis-gas chromatography/mass spectrometric (Py-GC/MS) analysis was carried out to identify the origin of the applied coating of the collected potsherds. ATR (attenuated total reflection) spectroscopy was used to confirm the application of organic substances on the surfaces of analysed potsherds.

To analyse a few samples, we used the Py-GC/MS method. With this method, a small amount (0.5 mg) of black coating can be scratched from the surface and analysed. Additionally, there is no need to destroy the whole sample, so it is an appropriate method for saving valuable archaeological materials. In contrast, the solvent extraction method requires a large amount samples for accurate analyses, and it is difficult to accurately analyse such small samples using this method.

Seven pieces of potsherds were selected for analysis using the Py-GC/MS method in the laboratory of the Department of Applied Chemistry at the School of Science and Technology at Meiji University in Tokyo, Japan. Four of these seven pieces are Type A, one is Type B, one is Type D and the remaining piece is Type E.

The Py-GC/MS measurements were carried out using a vertical micro furnace-type pyrolyser PY-2020id (Frontier Lab, Japan) and an Agilent 6890N/ 5975 GC/MS system (Agilent Tech. Santa Clara, CA). A stainless steel capillary
column (0.25 mm i.d × 30 m) coated with Ultra Alloy PY-1 (100% methyl silicone) was used for the separation. First, a small amount of sample (0.5 mg) was placed in tetra methyl ammonium hydroxide [TMAH] (1 µg). Next, the sample was pyrolysed by reactive pyrolysis gas chromatography/mass spectrometry analysis [TMAH/Py-GC/MS] in a thermal cracking furnace. A sample cup was placed on top of the pyrolyser at near ambient temperature. The sample cup was introduced into the furnace at 500ºC, and then the temperature program of the gas chromatograph oven was started. The gas chromatograph oven was programmed to provide a constant temperature increase of 20ºC per minute from 40 to 280ºC and then hold for 10 minutes at 280ºC. The injection and interface temperature was 280ºC, and the flow rate of helium gas was 1 ml/min. The mass spectrometry ionisation energy was 70eV (EI-mode).

Fourth, one piece of the black coloured potsherd of Type D was analysed using ATR spectroscopy. A thermo-Nicolet iN10MX FT-IR microscope fitted with an MCT detector cooled by liquid nitrogen was used. The ATR spectra were acquired within the range of 4000-650 cm⁻¹ at a spectral resolution of 4 cm⁻¹ using a slide-on ATR objective with glass. A background single-beam spectrum of the clean ATR crystal was first collected with the slide-on ATR objective inserted but not in contact with the sample surface. A total of 32 scans were recorded for interferogram averaging. For the ATR mapping on a selected area (420×110 µm for sample Type D), a step size of 20 µm in the x-direction and 10 µm in the y-direction was chosen with an aperture of 10.25 µm.

RESULTS

Observation of surface coating

Type A

All the collected potsherds of type A were observed by SEM and results showed that the place of high reflectivity of the electron image was a plain smooth coating. The place of low reflectivity of the electron image showed another coating on the smooth surface, and there were many cracks in it. Below the plain smooth surface, rough body clay was visible [Fig.6a].

A total of 200 glass slides were prepared from the collected potsherds to allow thin section observation. Thin section observation of Type A distinguished the presence of a two-stage coating on the body surface. Ground coat which is visible on the body surface is made from clay and
approximately 15-25 µm thick. Observation shows, the body clay and the clay particles of the ground coat are different. This ground coat is made from fine clay that is free from large mineral particles and uniformly applied to both the internal and external sides of the pottery. This refined clay made surface coat (ground coat) should be called as ‘slip’. The colour of the ground coat was deep brown. Over the ground coat was a black coloured top coat (2-4 µm thick) [Fig. 6b]. The thin section observation by SEM revealed numerous horizontal cracks in the thick ground coat. In contrast, the thin top coat exhibited numerous vertical cracks [Fig.6c].

Fig. 6 (a) Surface observation of the potsherds of Type A by SEM, body clay adjacent plain smooth coat (the place of high reflectivity) and top coat (the place of low reflectivity) is identified; (b) Thin section observation of Type A in optical microscope, thin black colour top coat is visible over the surface slip; (c) Observation by SEM, slip (1), top coat (2), cracks in top coat (3) is visible.

Type B
The surface of the collected potsherd of type B was observed by SEM and showed that the place of low reflectivity of electron image was a coating that contained many cracks [Fig. 7a].

In the thin sections of Type B had a black coloured coating (approximately
35 $40 \, \mu m$ in thickness) on the body surface that looked pure and non-clay-made [Fig.7b]. Many empty places created by bubbles were also found in the thin section observation. The body surface (below the coating) was not polished compared with Type A. In the polarization image by the optical microscope, a lump of reddish coloured clay was also identified below the surface coating. The body clay of the pottery was not especially fine, as many large mineral particles were located in the body surface. The SEM observation distinguished numerous cracks in the coating [Fig.7c].

**Fig. 7** (a) Surface observation of the potsherds of Type B by SEM, body clay (the place of high reflectivity) and surface coat (the place of low reflectivity) is identified; (b) Thin section observation of the type B potsherds in optical microscope, surface coat is visible; (c) Thin section observation by SEM, thick surface coat and body clay is visible.

**Type C**
The surface of the collected potsherds of type C were observed by SEM and showed that the place of high reflectivity of the electron image was a plain, smooth coating. The place of low reflectivity of the electron image showed another coating upon the smooth surface, and it had many cracks in it. Below the plain, smooth surface, rough body clay was visible [Fig. 8a]. The thin sections of Type C indicated that a two-stage coating was present on the
surface of the potsherd. The ground coat or slip is made from fine clay and is 25-30 \( \mu m \) thick. A thin coat was identified over this ground coat [Fig.8b]. The body clay of the analysed potsherds was fine, and large mineral particles were not identified. The SEM observation revealed horizontal cracks in the ground coat and vertical cracks in the top coat.

**Type D**

Surface of the collected potsherd of type D was observed by SEM and showed that the place of low reflectivity of electron image was a multi-staged coating upon the body clay, and it had many cracks in it [Fig. 9a].

The thin section observation of Type D revealed a four-staged dark red coating on the body clay, and it did not contain any clay material like the ground coating of Type A. The thickness of the coating was approximately 25-30 \( \mu m \) [Fig. 9b]. The body clay contained many large mineral particles. The SEM observation located numerous cracks in the coating [Fig. 9c].
Fig. 9. (a) Surface observation of the potsherds of Type D by SEM, body clay (the place of low reflectivity) and adjacent multi-staged surface coat (the place of high reflectivity) is identified; (b) Thin section observation of the type D potsherds in optical microscope, body clay, thick and multi-staged surface coat is identified; (c) Thin section observation of the potsherd of type D by SEM, body clay, thick surface coating and cracks of the coating are visible.

Type E
Surface of the collected potsherd of type E was observed by SEM and showed that the place of low reflectivity of electron image was a coating upon the body clay with many cracks in it [Fig. 10a]. The cracks of this coating were similar to Type B and Type D. In the thin section observation of Type E, a one-staged coating was identified on the surface that was approximately 25 – 30 µm thick [Fig. 10b]. The SEM observation revealed vertical cracks in the coating, different from Type B and Type D [Fig. 10c].
Fig. 10. (a) Surface observation of the potsherds of Type E by SEM, body clay (the place of high reflectivity) and adjacent surface coating (the place of low reflectivity) is identified; (b) Thin section observation of the type E potsherds in optical microscope, body clay and surface coating is identified; (c) Thin section observation of the potsherd of type E by SEM, body clay, surface coating and vertical cracks in the coating are visible.

Py-GC/MS Analysis
The analysis results of the seven analysed potsherd samples are summarised in Table 1.

Table 1. List of the black coloured potsherd samples used for pyrolysis and analysis results.

<table>
<thead>
<tr>
<th>Analyzed potsherd samples</th>
<th>Collection Method</th>
<th>Analysis results</th>
</tr>
</thead>
<tbody>
<tr>
<td>No.1 Type A Scratched</td>
<td></td>
<td>Methyl palmitate and Methyl stearate</td>
</tr>
<tr>
<td>No.2 Type A Scratched</td>
<td></td>
<td>Methyl palmitate and Methyl stearate</td>
</tr>
<tr>
<td>No.3 Type B Scratched</td>
<td></td>
<td>Methyl palmitate and Methyl stearate</td>
</tr>
<tr>
<td>No.4 Type C Scratched</td>
<td></td>
<td>Methyl palmitate and Methyl stearate</td>
</tr>
<tr>
<td>No.5 Type C Scratched</td>
<td></td>
<td>Methyl palmitate and Methyl stearate</td>
</tr>
<tr>
<td>No.6 Type D Scratched</td>
<td></td>
<td>Methyl palmitate and Methyl stearate</td>
</tr>
<tr>
<td>No.7 Type E Scratched</td>
<td></td>
<td>Methyl palmitate and Methyl stearate</td>
</tr>
</tbody>
</table>

The TIC and mass chromatogram ($m/z = 74$) of Type A are shown in Fig. 11, and the mass spectra analysis is shown in Fig. 12. The results show that the retention time of 18.24 min is methyl palmitate (peak 1) and at 19.84 min is methyl stearate (peak 2).

The results of the mass chromatogram ($m/z = 74, m/z = 151$) for Type B, Type D and Type E are shown in Fig. 13. The names of the peaks that are shown in Fig. 13 are listed in Table 2 and Table 3. Methyl palmitate and methyl stearate were detected in the mass spectrum (Figs. 14 and 15). Moreover, from Fig. 17, the mass chromatograph of peak 3 ($m/z = 382$) shows the substance 3-(12-phenyl)dodecyl catechol and Mass spectra of standard 3-(12-phenyldodecacyl) catechol dimethyl ether (M.W-382) is shown in Fig. 18.
Fig. 11. *The TIC and mass chromatogram (m/z = 74) of Type A potsherds.*

Fig. 12. *The mass spectra (Type A potsherds) of peak 1 and peak 2; Peak 1: Methyl palmitate, Peak 2: Methyl stearate.*
Fig. 13. Mass chromatogram ($m/z = 74$, $m/z = 151$) of the black coloured potsherds of Type B, Type D and Type E.

Table 2. List of the peaks of mass chromatogram ($m/z = 74$) (Type B, Type D and Type E).

<table>
<thead>
<tr>
<th>m/z 74</th>
<th>Name of the peaks and structure</th>
</tr>
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<tbody>
<tr>
<td>Peak 1</td>
<td>Methyl hexanoate</td>
</tr>
<tr>
<td>Peak 2</td>
<td>Methyl heptanoate</td>
</tr>
<tr>
<td>Peak</td>
<td>Molecule</td>
</tr>
<tr>
<td>-------</td>
<td>-------------------</td>
</tr>
<tr>
<td>3</td>
<td>Methyl nonanoate</td>
</tr>
<tr>
<td>4</td>
<td>Methyl decanoate</td>
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</tr>
<tr>
<td>10</td>
<td>Methyl palmitate</td>
</tr>
<tr>
<td>11</td>
<td>Methyl stearate</td>
</tr>
</tbody>
</table>
Table 3. List of the peaks of mass chromatogram (m/z 151) [Type B, Type D, and Type E].

<table>
<thead>
<tr>
<th>Peak</th>
<th>Name of the peaks and structure</th>
</tr>
</thead>
<tbody>
<tr>
<td>Peak 1</td>
<td>Methyl hexadec-9-enoate (Palmitoleic acid methyl ester)</td>
</tr>
<tr>
<td>Peak 2</td>
<td>Methyl octadec-9-enoate (Oleic acid methyl ester)</td>
</tr>
<tr>
<td>Peak 3</td>
<td>3-(12-phenyl-dodecacyl)catechol</td>
</tr>
</tbody>
</table>
Fig. 14. Mass spectra of peak 10 in Fig. 13 and standard methyl palmitate (Type E potsherd).

Fig. 15. Mass spectra of peak 11 in Fig. 13 and standard methyl stearate (Type E potsherd).
Fig. 16. Mass chromatogram (m/z = 382) of the black coloured potsherd of Type E.

Fig. 17. Comparison of retention time (25.23 min.) time of the mass chromatography (m/z = 382) of the black coloured potsherd of Type E and standard 3-(12-phenyldecacyl) catechol dimethyl ether in the lacquer film of Melanorrhoea usitata (Myanmar).
Fig. 18. Mass spectra of standard 3-(12-phenyldodecacyl) catechol dimethyl ether (M.W. 382).

**ATR mapping**

The ATR mapping result of a measurement of Type D is shown in Figs. 19-21 and Table 4. In Figure 19, 1700 cm\(^{-1}\) is the peak of a C=O bond of organic acid, and there was stronger absorption in the red part. In contrast, a blue part had weak absorption. The ATR mapping showed that a component with a C=O bond exists at high concentration on the surface of the analysed potsherd.
Fig. 19. Measuring range of ATR (Inner side of a black line).

Fig. 20. A profile of a C=O bond (1700 cm⁻¹).

Fig. 21. IR spectra of the parts of a white X point.
Table 4. List of wave number and bond types

<table>
<thead>
<tr>
<th>Wave number</th>
<th>Bond Type</th>
</tr>
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<td>2919</td>
<td>ν (C−C)</td>
</tr>
<tr>
<td>1700</td>
<td>ν (C=O)</td>
</tr>
<tr>
<td>1565</td>
<td>ν (C=C)</td>
</tr>
<tr>
<td>1357</td>
<td>δ (C−H)</td>
</tr>
</tbody>
</table>

DISCUSSION

The microscopic observation suggests that all 30 pieces of the potsherds of black coloured pottery had a thick coating on their surfaces. The coatings of the potsherds of Type A and Type C were applied with same technology, and they contained a two-stage layer with two different origins. These potsherds of Type A and Type B are the original potsherds of NBPW. Previous researcher like Robert Harding’s research (2004) concludes that ‘all the examined samples have two slips, with the upper surface being approximately a micron in width…’22. Present research has identified that the ground coat was made from clay (called ‘slip’) and was approximately 15~35 µm thick. However, clay was not identified in the top coating, which is 2~4 µm thick. From the result of Py-GC/MS analysis, it is confirmed that potsherds of Type A and Type C contained oil (fatty acid methyl esters) on their top coating. So, the slip was applied for achieve the black colour of the surface and top coating was for the lustrous black surface of NBPW.

The analysed black coloured potsherds of Type B, Type D and Type E were different from the potsherds of Type A and Type C. The thin section observation identified a solid and thick coating on the surface of the potsherds of Type B, Type D and Type E. This solid coating was applied directly on the body surface of the pottery, and no clay-made slip was present, like with Type A and Type C. Compared with Type A and Type C (NBPW), the black coloured potsherds of Type B, Type D and Type E are different with respect to the coating technique.

In the potsherd of Type B, the thin section observation identified a thick coating (approximately 35~40 µm) and many empty round spaces inside the surface.

coating. This characteristic may indicate that organic material was applied to the warm surface condition of the pottery (approximately 100°C) and that the surface temperature of the pottery created the bubbles in the coating. The thin section observations also suggested that coating was applied over a rough surface of the pottery, that’s why the applied coating is not uniform. From the thin section observation, this type was completely different from Type A and Type C.

In the Type D potsherd, the surface coating consisted of 4 layers and was approximately 25–40 µm thick. The visual surface of this potsherd was black and without glossiness. The multi-staged coating technique was supposed to obtain the black colour. In the potsherd of Type E, which was approximately 25–30 µm thick, a one-staged and solid coating was found in the thin section observation. The thin section observation in SEM revealed vertical cracks in the coating, similar to the cracks of Type A and Type C (top coat).

The Pyrolysis-Gas Chromatography/Mass Spectrometric analysis identified ‘oil’ (fatty acid methyl esters) in the surface coating of the collected black coloured potsherds of Type B and Type D, and the substance 3-(12-phenyldodecyl) catechol was also identified in the coating of Type E.

The ATR mapping analysis of the potsherd from Mahasthangarh site (Type D) was carried out to confirm the application process and showed that a component with a C=O bond exists in high concentration on the surface of the analysed potsherd. In case organic acid sank in the pottery from the surrounding ground when it was buried, the bond of C=O should exist uniformly to the inside of the body clay. However, the ATR mapping data indicate that the coating was uniformly condensed on the surface of the examined earthenware (Type D). On the basis of this measurement result, there is a possibility that oil was applied as a coating material over the surface of the analysed potsherd and that this applied coating was not any type of contamination. The image of thin section produced by the transmitted light micrograph also clearly showed the smooth layers on the pottery surface.

We knew from the previous research of B.P. Sinha that ‘black ware’ and ‘burnished black (black polished) ware’ was also common in the NBPW level of Sonpur, located on the east coast of India.23 He mentioned that ‘some sherd had a

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wash on their exterior surface, but whether the slip is completely peeled off or the ware is black without slip is difficult to ascertain. In the present research, analysed potsherd (Type E) from the Mahasthangarh site possessed a similar quality with those Sonpur specimens; it contained a coating in a small portion of the interior side that was also extremely fragile [Fig. 5c].

The substance 3-(12-phenyldodecyl) catechol was identified in the surface coating of Type E by Py-GC/MS analysis. This substance is considered to have originated from thitsiol, which is the main ingredient of *Melanorrhoea usitata* (one kind of lacquer harvesting tree). In this study, the lacquer film (made from the tree sap of *Melanorrhoea usitata*, collected from Myanmar) was analysed for a comparison between the surface coating of Type E potsherd and the original tree sap of *Melanorrhoea usitata*. A substance with the same mass spectrum was found in the lacquer film of *Melanorrhoea usitata* and Type E, and it was measured in the retention time for 25.23 minutes. Therefore, this potsherd sample (Type E) also contained the same lacquer sap as *Melanorrhoea usitata*. The amount of 3-(12-phenyldodecyl) catechol substance in the analysed potsherd was very low compared with the fatty acid methyl ester substance.

Thitsiol components such as 3-(12-phenyldodecyl) catechol cannot be detected from the top coat of NBPW (Type A and Type C). So, by the analysis of Py-GC/MS, not only oil can distinguish but discriminate between oil and other natural resins also.

Further research about the lacquer will be carried out by analysing the large amount of potsherd samples of Bangladesh, which may deliver some new information. Myanmar, one of the countries neighbouring Bangladesh, also possess lacquer culture from their past to present. Thus, it is assumed that the lacquer culture from Myanmar was exercised in the ancient pottery of Bangladesh. Lacquer has been used in Asian countries for thousands of years as a durable and beautiful coating material. Three kinds of lacquer trees grow in the East: *Rhus vernicifera* (Japan, China and Korea), *Rhus succedanea* (Vietnam), and *Melanorrhoea usitata* (Thailand and Myanmar). The phenol derivative of *R. vernicifera* is urushiol, of *R. succedanea* is laccol and of *M. usitata* is thitsiol.

24 Ibid.

In Bangladesh, several types of resinous tree are present, for example, Sal tree (*Shorea robustus*, tree sap: Dammar), Neem tree (*Azadirachta indica*, tree sap: Neem gum), Mango tree (*Magnifera indica*, tree sap: Mangiferin), and Udal tree (*Sterculia villosa* Roxb, tree sap: Gum karaya). However, lacquer harvested trees are not available presently in Bangladesh. So, the presence of lacquer coated pottery in present Bangladesh could suggest us two ideas,

First, lacquer harvested tree was present in ancient Bangladesh, and harvested lacquer was used in the indigenous pottery for surface coating.

Second, lacquer was imported from outside of Bangladesh and used as a coating material of the surface of indigenous pottery.

Third, Lacquer coated pottery was imported from outside of Bangladesh.

Usually, drying oil (for example, pine tar, perilla oil, or linseed oil) is used in lacquer as a thinner, which is then applied on the surface of the different types of artistic materials. Lacquer is expensive and difficult to obtain because the harvesting process is costly. It is therefore suggested that potters of those ancient days may have experimented with different types of resin oil-made and solely oil-made surface coating. Another possibility is that different types of pottery may have been used for the people of different social classes on the basis of their ranking and that the analysed potsherds were some of those items.

**Conclusion**

In this study, 30 pieces of black coloured potsherds were examined that were excavated from two early historic sites in Bangladesh. Among the collected potsherd samples, three different types are distinguished by the observation of thin sections. The NBPW is one of these types; the other two are totally new types of potsherds. A clay-made ground coat (like that of NBPW) was not identified in these new types of potsherds. The results of Py-GC/MS led to the conclusion that oil (fatty acid methyl ester) and thitsiol [3-(12-phenyl-dodecacyl) catechol] are present in the surface coating of these potsherds. This study is a preliminary source for investigating the manufacturing technique of the black coloured ancient pottery of Bangladesh as well as South Asia. The present study claims that NBPW possess two different types of coating which was originated from two different source and tree based lacquer coated pottery was used in ancient Bangladesh.