Mineralogical characterization of raw materials from Dholavira, Gujarat, India and its geological and archaeological significance

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Dholavira was one among the five largest settlements (probably six) of the Harappan civilization during the third millennium BCE. The location of this site in a desolate corner of Khadir Island in Kachchh, Gujarat, India, speaks well of the planning and ingenuity showed by the Harappans for making it an urban and administrative centre. Excavations at this site between 1989 and 2005 brought to light a long and continuous occupation of nearly 1500 years (*c.* 3000–1500 BCE), which records the rise, culmination and fall of the Harappan Civilization. Evidences for various craft activities are found from the earliest levels onwards and the Harappans exploited various lithic raw materials for both utilitarian purposes and making ornaments. Kachhch and the Gujarat mainland are rich in raw material resources in general and agate–carnelian, limestone, various types of clay, copper–lead–silver and steatite, in particular. Dholavira contains both raw materials and finished artefacts, thus presents an ideal scenario to study. In the present study, we interpret the mineralogical characterization and probable provenance of the raw materials from different spatio-temporal contexts at the Dholavira site using techniques like XRD and SEM-EDS analysis. Samples of clay, stone raw materials and a few artefacts were selected from among the innumerable resources available at the site.

Keywords: Archaeological site, artefacts, geological framework, mineralogical characterization, raw materials.

THE study of prehistoric exchange systems, reconstruction of trade and source, and understanding the economic aspects of ancient societies are perhaps among the major topics in archaeology[1–7]. The long-drawn processes of settled human life along with the domestication of plants and animals, technological innovations and long-distance trade have been well documented from a host of sites in South Asia, starting from the Neolithic site of Mehrgarh in Pakistan[8]. The sourcing, procuring, processing and distribution of the raw materials from diverse sources during different phases of the Harappan Civilization illustrate the emergence of state-level society during the third millennium BCE[6]. Aspects related to material characterization, technology and long-distance exploitation of raw materials have been a matter of interest among scholars right from the discovery of the Harappan Civilization[4,6,9–12].

More sophisticated instrumentation like X-ray diffraction (XRD), scanning electron microscopy (SEM), energy dispersive X-ray spectroscopy (EDS), electron probe microanalyser (EPMA), neutron activation analysis (NAA), thermal ionization mass spectrometer (TIMS) and inductively coupled plasma mass spectrometry (ICP-MS) are now commonly used for mineralogical, elemental and isotopic composition as tracers for the sources of raw materials[5,13,14].

In the present study, using techniques like XRD and SEM-EDS, we have analysed major raw materials from the Harappan site of Dholavira in the Kachchh district of Gujarat, India. XRD analysis of clay and lithic samples reflects their mineralogical characterization and SEM-EDS has been used for the morphological, structural and chemical assessment of the lead nodule and the bangle fragment. The main aim of this study is to develop a mineralogical database to characterize and interpret the expected provenance and archaeological origins, that would be crucial for understanding the Harappan network of resource acquisition and inter-regional interactions[6]. Understanding these various categories of materials will be crucial for the complex processes of raw material acquisition and exploitation during the third millennium BCE[2,3,5,7,13–16].

Dholavira Harappan site

Archaeological framework

Dholavira (locally known as Kotada Timbo), a UNESCO world heritage archaeological site of the Harappan Civilization (*c.* 2600–1900 BCE), is located on a small isolated island of Khadir bet, Bhachau taluka in the salt plains of Rann of Kachchh in the Kachchh Peninsula, northern Gujarat. The contemporary non-urban phase known as Sorath Harappan is well represented from the sites in the Saurashtra Peninsula, while sites of the Harappan phase are mostly located in the Kachchh peninsula (Figure 1). It was excavated by the Archaeological Survey of India from 1989–90 to 2004–05 under the direction of R. S. Bisht,

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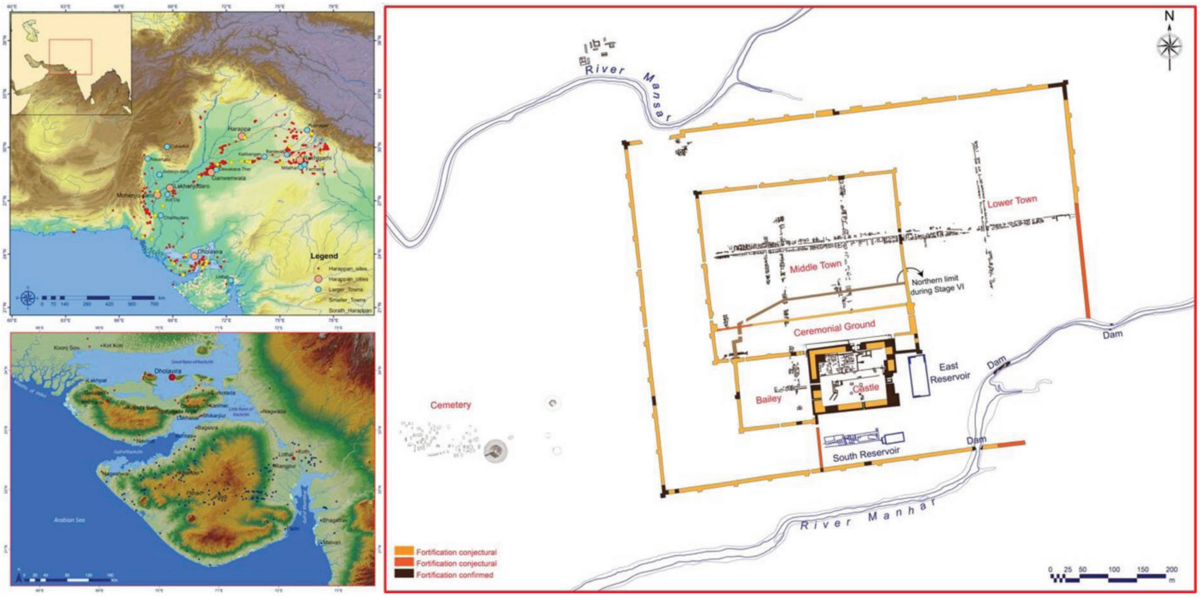

Figure 1. Layout view of different divisions of the Dholavira Archaeological Site, Gujarat, India (Right). The inset maps showing the various Harappan sites (top left) and Gujarat Harappan sites (bottom left).

observed according to whom Dholavira has indeed added new dimensions to the personality of the Harappan Civilization[1,2,17]. The settlement was under occupation for nearly 1500 years, starting from around 3000 BCE, witnessing several stages of cultural evolution and amalgamation, growth and decline[17]. The ruins cover an area of 100 ha between two seasonal channels named Manhar and Mansar, which were harnessed to fill a series of water reservoirs within the walled city (Figure 1). The hallmark findings from the settlement include a meticulously laid-out urban settlement with at least three areas individually fortified and a fourth area within the overall outer fortification; water management and harnessing systems, aesthetic pillar elements, long-distance contacts with Sindh and Punjab regions, craft activities on various mediums such as shell, chert, agate-carnelian, stoneware, copper and gold, to name a few. The unique funerary architecture at Dholavira indicates the presence of a distinct socio-economic group with diverse belief systems[17].

Geological framework

Geologically, Dholavira lies in the Khadir Island, one of the island belts consisting of four discontinuous land masses, i.e. Pachham, Khadir, Bela and Chorar islands, progressively from west to east with Wagad in the south[18]. These regions are mainly covered by Mesozoic and Tertiary formations. Lithologically, the Mesozoic successions of the Jhurio Formation consist of a thick sequence of limestone and shales with different bands of coloured limestone, which lies in the northern boundary of Khadir. The overlying formation of Jumara, named after Jumara hill near the Rann, is mainly argillaceous shales laminated with monotonous olive-grey gypseous with thin ferruginous bands. Jhurio formation consists of three members – lower, middle and upper. The lower member consists of red sandstone and shale and the middle is dark-grey laminated gypseous shale. The upper member of Jharun consists of massive sandstone beds with intercalations of shales. Another important geologic formation is the Bhuj Formation, exposed beside the Jhumra Formation in the south. Some exposures are the outer boundaries of Wagad Island. Massive and uniform non-marine sandstone prevails in this Formation. The tertiary formation overlying the Deccan trap covers mostly the western and southwestern part of Kachchh with three major series, namely Berwali, Bermoti and Khali, from older to younger sequences respectively, from Eocene, Oligocene and Miocene[19]. The quaternary succession includes mostly Rann sediments (salt marsh), alluvial fans and many coastal deposits[18].

The rich geological deposits of Kachchh were interlaced with several archaeological sites, which were strategically located to harness the mineral-rich resources nearby. The surrounding lithology around Dholavira has wide and varied lithological sources, which the Harappans might have used for different purposes. Therefore, studying of various raw materials and artefacts excavated from the Harappan sites is important for understanding their geological and archaeological provenances.

Materials and methods

Macroscopic examination

This is the simplest, cheapest and yet most prominent method to identify raw materials. The macroscopic examinations are helpful to a great extent in identifying the state of origin of raw materials, i.e. igneous, sedimentary or metamorphic phases with a preliminary inference of minerals, etc. In this study, a total of 32 raw materials from Dholavira sampled from different phases were chosen (Figure 2 *a* and *b*). One sample of ernestite (IITGN-E1 and IITGN-E2) (Figure 2 *b*) from Bhagatrav, a Sorath Harappan site, was also analysed. The major raw materials from Dholavira were either clay type or lithic/stones. One sample each of lead nodule (metallic) and faience bangle fragment was also characterized and analysed for SEM and energy-dispersive X-ray spectroscopy (EDS) analysis (Figure 3). The selected samples from Dholavira have been tabulated in terms of major clay type (Table 1) and lithic type samples (Table 2). Figure 4 *a* and *b* shows their approximate locations on the layout map of the Dholavira site. The samples of lithic raw materials were identified as igneous rocks/minerals (different varieties of quartz), jasper, chert, bead fragments, metallic nodules, bangle fragments, etc.

XRD analysis

XRD analysis enables to determine the various mineral (crystalline) phases present in a sample. This is the primary technique used to determine the physico-chemical make-up of unknown solids. The sample to be analysed is finely powdered and homogenized. XRD analysis is also useful to determine the polymorphs of a mineral, and hence pressure–temperature conditions for the formation of materials can be interpreted. X-ray diffractograms can also be evaluated for the relative proportion of the identified mineral phases. XRD analysis of Dholavira samples was done using a Bruker D8 Advance X-ray system with CuKα radiation and monochrometer at 40 kV/40 mA with scans run at 5°–80° (2θ) having 0.02° step size and 0.25 sec count time at the Indian Institute of Technology, Gandhinagar (IITGN).

Figure 2. Photographs of raw materials from the Dholavira. (***a***) Samples of clay-type raw materials and (***b***) samples of lithic raw materials.

Figure 3. Raw materials with ornamental significance from Dholavira: the metallic lead nodule (A1 and A2) and a faience bangle fragment (B-1 and B-2).

Table 1. Location and description of clay samples from Dholavira archaeological site, Gujarat, India

Sample	Locus (m × m × m)	Stratum	Depth (m)	Material (inferred)
IITGN-C1	37 × 76 × 2	4	–2	Red oxide
IITGN-C2	–	–	–	Clay
IITGN-C3	37 × 77 × 1	3	55	Clay
IITGN-C4	37 × 78 × 2	2	–21	Clay
IITGN-C5	37 × 78 × 3	Surface	–	Clay
IITGN-C6	47 × 33 × 3	4	50	Chalk
IITGN-C7	48 × 9 × 1	3	–90	Clay
IITGN-C8	47 × 85 × 4	2,7	–105	Clay stone
IITGN-C9	37 × 79 × 3	1	140	Chalk stone
IITGN-C10	45 × 43 × 2	10	–156	Plaster of Paris
IITGN-C11	H19/1&4	2	–	Chalk stone
IITGN-C12	47 × 83 × 2	pit	–178	Clay stone
IITGN-C14	–	–	–	Clay

Table 2. Location and description of lithic samples from Dholavira archaeological site

Sample	Locus (m × m × m)	Stratum	Depth (m)	Material (inferred)
IITGN-S2	37 × 79 × 1	1	60	Terracotta bead fragment
IITGN-S7	48 × 88	6	–185	Soapstone
IITGN-S13	37 × 58 × 3	12	–505	Cherty haematite
IITGN-S14	35 × 73	3	–90	Chert stone
IITGN-S20	38 × 94 × 3	2	–345	Slag (malachite)
IITGN-S21	55 × 52	2	55	Feldspar
IITGN-S26	Eastern reservoir	–	Surface	Chert (quartz)
IITGN-S36	37 × 76 × 4	2	52	Stone
IITGN-S39	47 × 85 × 1	6	–70	Bead stone
IITGN-S40	47 × 95 × 2	–	–102	Stone
IITGN-S42	47 × 85 × 4	8	–124	Ochre stone
IITGN-S54	37 × 76 × 4	2	46	Soapstone?
IITGN-S65	47 × 84 × 3	5	–155	Ochre stone
IITGN-S67	54 × 68 × 1	19	–280	Dark stone
IITGN-S86	47 × 35 × 4	3 (s.b.8)	–123	Stone
IITGN-S112	25 × 2 × 4	2	–31	Stone (quartz)

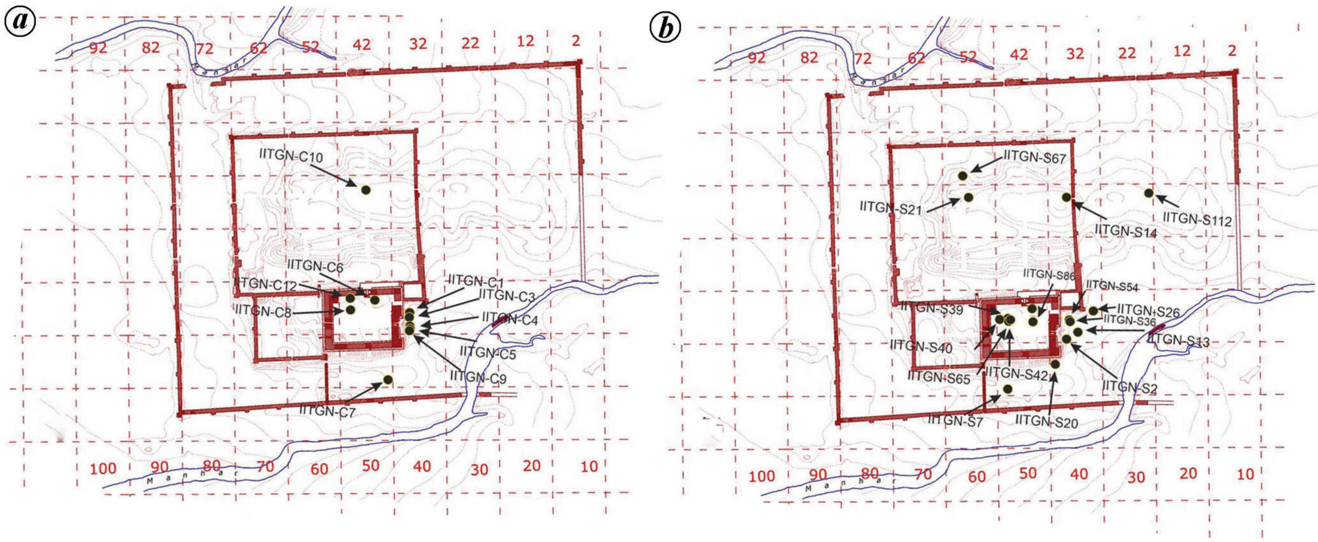

Figure 4. Sample location on the layout view of Dholavira archaeological site for (***a***) clay samples and (***b***) rocks and lithic fragments.

Semi-quantitative calculations of each peak were carried out using the Bruker EVA 3.0 program. The relative abundance of various minerals was estimated using the particular peak area ratio of each identified mineral on the EG diffractogram.

We had chosen 30 samples for XRD analysis, of which 13 were of clay variety (Table 1) while the remaining were lithic (Table 2). Further, two fragments of ernestite from Bhagatrav were also analysed.

SEM-EDS analysis

SEM is a strong imaging technique to study the micro-textures and their compositions. EDS gives the quantitative analysis of the various compositions. This is a powerful tool for the analysis of artefacts. It can give a magnified view of the samples up to a few nanometers. The EDS spectrum provides an elemental analysis of the sample. The advantage of EDS is that the chemical characterization of the sample at any spatial location in the SEM image can be obtained. The energy of the characteristic X-rays allows the detection of the element of the parent atom. Thus, SEM is a useful tool for elemental mapping with a qualitative evaluation of the composition of the examined area. Two samples, viz. a lead nodule and bangle fragment, were chosen for SEM-EDS analysis.

Results and discussion

XRD results and interpretation

Thirteen clay samples were analysed for their mineralogical composition using XRD. Most of the clay samples contained kaolinite, while some samples like IITGN-C3, IITGN-C4,

Table 3. Relative proportion of mineralogical composition of clay samples from Dholavira from XRD analysis

Sample	Quartz	Kaolinite	Smectite	Illite	Haematite	Tridymite	Gypsum	Apatite	Apophyllite	Zeolites	Dolomite	Figure
IITGN-C1	11.06		59.09		25.1	4.75						9 *a*
IITGN-C2	31.4	60.36				8.23						9 *c*
IITGN-C3							25.57	52.45	22			9 *b*
IITGN-C4	75.35			17.38	1.71			5.57				9 *d*
IITGN-C5	6.7	61.2	32.1									9 *e*
IITGN-C6		29.46	20.57		7.24	17.57		25.14				9 *f*
IITGN-C7	6.2	69.6						24.17				9 *g*
IITGN-C8	3	76.5								20		9 *h*
IITGN-C9		78.1					21.8					9 *i*
IITGN-C10						7.8	85.2				6.9	9 *j*
IITGN-C11	11.2	12.5		75.2								9 *k*
IITGN-C12		54.41	10.2			9.2		26.1				9 *l*
IITGN-C14	15	61	23									10 *a*

Table 4. The major and minor mineral composition of lithic samples from Dholavira through XRD analysis

Sample	Material (inferred)	Major minerals (XRD analysis)	Minor minerals (XRD analysis)	Figure
IITGN-S2	Terracotta bead fragment	Quartz	Goethite, Kaolinite	10 *c*
IITGN-S7	Soapstone	Chlorite, talc		10 *b*
IITGN-S13	Cherty haematite	Haematite, smectite	Quartz	10 *d*
IITGN-S14	Chert stone	Not analysed		
IITGN-S20	Slag (malachite)	Cu_2CO_3, $CaCO_3$	Hercynite, sillimanaite	10 *e*
IITGN-S21	Feldspar	Microcline, anorthoclase	Quartz	10 *g*
IITGN-S26	Chert (quartz)	Not analysed		
IITGN-S36	Stone	Smectite, quartz	Goethite	10 *f*
IITGN-S39	Bead stone (agate)	Not analysed		
IITGN-S40	Stone	Albite, goethite	Quartz	10 *h*
IITGN-S42	Ochre stone	Heamatite, goethite	Serpentine, quartz	10 *i*
IITGN-S54	Soapstone?	Kaolinite		10 *k*
IITGN-S65	Ochre stone	Haematite	Quartz	10 *j*
IITGN-S67	Dark stone	Not analysed		
IITGN-S86	Stone	Quartz		10 *l*
IITGN-S112	Stone (quartz)	Not analysed		

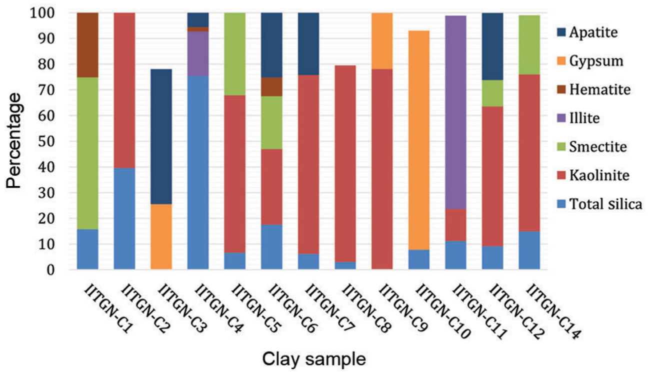

Figure 5. Mineralogical composition of clay samples from Dholavira.

IITGN-C10 and IITGN-C11 dominated apatite, silica, gypsum and illite clay (Figure 5 and Table 3). The majority of the samples had smectite (montmorillonite) and kaolinite; a few samples contained a high proportion of apatite, while illite and gypsum were present in one or two samples in significant proportions. The basic minerals identified in the samples were illite, kaolinite and quartz, haematite (red clay), gypsum (chalk stone) and apatite (Tables 3 and 4).

Major lithic materials identified were different varieties of quartz like chert, agate, amethyst, jasper, chalcedony, carnelian, etc. A few other silicate minerals like amazonite and microcline were also identified. Some of the raw materials were recognized as sedimentary rocks like sandstone, siltstone, gypsum and limestone. The third category of samples were rich, such as malachite, haematite, lead (galena) and ochre-stone (Table 4). The characteristics of a few important lithic raw materials identified from the Dholavira samples are detailed below.

Chert: This was observed as one of the most abundant excavated raw materials of Dholavira. It has been defined as opaque microcrystalline silicate having a colour that is either neutral (shade of black/white) or a shade of brown (as defined in Law[6]). Chert is a sedimentary rock consisting entirely of silica (SiO_2) formed due to the precipitation of SiO_2-rich liquid. Samples like IITGN-S14 and IITGN-S26 are typical examples of chert (Figure 2 *b*). They are usually identified through hand specimens. Cherts were mostly utilized to make blades, cutting tools, scrapers, etc.

Agate and jasper/chalcedony: These minerals are also found abundantly in Dholavira. They are crypto-crystalline varieties of quartz, usually having gem quality. Samples IITGN-S39 and IITGN-S112 are typical examples (Figure 2 *b*). They are fundamental materials for making beads and jewellery, along with standardized cubical stone weights[14]. Dholavira has been identified as a major bead-manufacturing centre utilizing various stones of silicious materials along with the enigmatic 'ernestite' drills[16].

Feldspar: Various types of feldspar were identified using XRD analysis, e.g. microcline and anorthoclase. Sample IITGN-S21 is a typical example of amazonite consisting of major minerals such as microcline with minor amounts of anorthoclase and quartz (Figure 2 *b*). They have a typical greenish tint and were used for making gem stones by Harappans. Several complete and unfinished beads of amazonite have been found at Dholavira. A few of them were broken exactly in the middle, which may be due to the crystalline structure and pressure exerted during the drilling mechanism.

Ochre minerals: These can be categorized as sedimentary rocks having a distinguishing red colour and consisting of sand/silt-sized grains. XRD analysis revealed that the major phases of the materials were Fe-rich carbonates, haematite and quartz, e.g. samples IITGN-S12 and IITGN-S65 (Figure 2 *b*). The Harappans mostly used them for artistic designs on ceramics. Ochres have also been used as mineral pigments during different periods in the past[20]. The ceramics from Dholavira are a combination of typical Harappan types as well as local varieties, which continued from the preceding early Harappan phase, with minor modifications[17]. A distinctive slip applied to the unbaked ceramics turns more reddish after firing. The red-coloured finish was obtained after heating in a furnace to around 900°C.

Sandstone/siltstone: These were abundantly found at the site and mainly exploited for fashioning stone blocks of different sizes for construction. A few of the sandstones having colouration properties have also been used for making perforating beads (e.g. terracotta beads, IITGN-S2) (Figure 2 *b*).

Cu and Pb ore fragments: There were a few metallic oxidized ores, e.g. Cu and Pb ores. XRD analysis of such materials, e.g. IITGN-S20 (Figure 2 *b*), revealed that they were mostly carbonates of Cu with a minor presence of $CaCO_3$. Various other minor minerals like hercynite and sillimanite were also observed. Bisht *et al.*[21] also highlighted the role of Dholavira in utilizing ores from various raw material resources.

Drill bits: Another significant raw material studied was ernestite from Bhagatrav of the Sorath Harrapan phase. This was basically used as a drill bit for perforating beads of the quartz family[14,16,17]. The geological provenance of this raw material is still untraceable even after studies spanning nearly a century[6]. XRD analysis was done for two fragments of ernestite nodules, on the black and grey-coloured portions separately (IITGN-E1 and E2 respectively). The black part of ernestite was like inclusion in grey country rock. Other features like mineral zoning were also evident, possibly due to the hydrothermal intrusion during metasomatic transformation. The minerals in the ernestite black fragments were sillimanite, haematite, mullite and quartz. The grey portions of ernestite had significant amounts of quartz, sillimanite and mullite. Sillimanite and mullite are aluminium silicates considered typical metamorphic minerals (Figure 6). The texture of ernestite showed possibly metasomatic transformations and interlocked structures, which add to its strength, thus making it suitable for use as a drill bit by the Harappans. The presence of mineral phases such as haematite, quartz, sillimanite and mullite in the ernestite nodules at Bhagatrav resembles the XRD results of drill bit materials from Dholavira[16,22]. The presence of ernestite from the Sorath Harappan site of Bhagatrav indicates its popular use for bead manufacturing even in the peripheral regions of Harappan culture. Ernestite was preferred by the Harappans for perforating

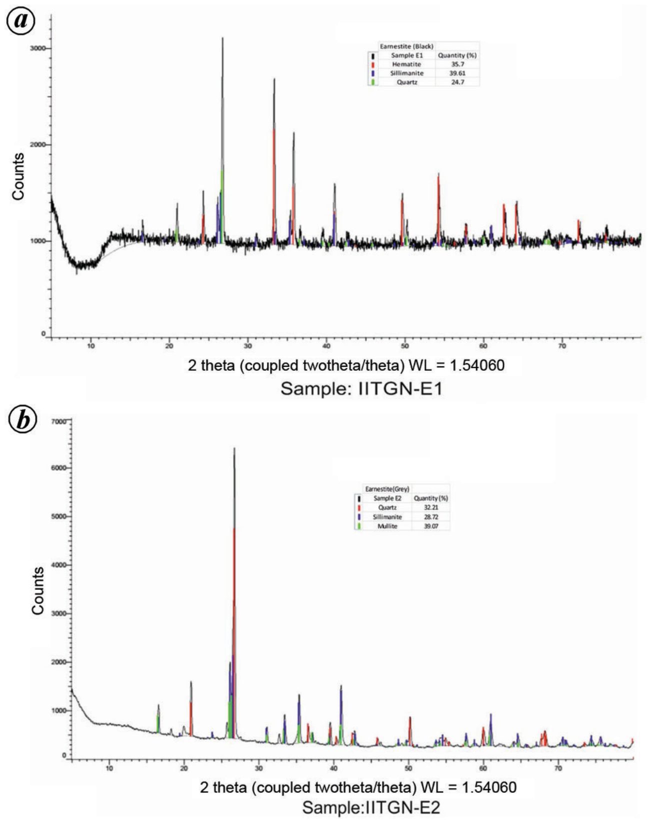

Figure 6. XRD pattern of 'ernestite' nodule with black (IITGN-E1) fractions (top) and grey (IITGN-E2) fractions (bottom).

harder stones of agate and carnelian due to its chipping properties and a smooth polished interior finish.

SEM-EDS results and interpretation

The lead nodule: The SEM-EDS scans were carried out at multiple points on the lead nodules. These scans revealed that the whole nodule (95–98%) is made of lead (Pb) and not anticipated galena (PbS), as there was no prominent sulphur peak in the EDS graph (Figure 7 *a*). A few minor peaks of Pb, As and C were evident in the scans. However, the exact percentage of arsenic (As) could not be detected in them. One of the scans of the nodule also showed peaks of O and Fe, but their percentage was found to be below the detection limit. Since the lead nodule contained over 95% lead, it was probably smelted or extracted Pb. A few lead ornaments have been excavated from Dholavira, which indicates the exploitation of this metal apart from using galena as surma, a popular eyeliner during ancient times. In this context, a burial from Harappan yielded evidence of a surma container[6], which substantiates the use of galena as an eyeliner by the Harappans.

Faience bangle fragment: The SEM-EDS analysis of the bangle fragment was aimed to identify its micro-texture (grain size and morphology) and variations in chemical composition from its outer, middle and inner layers. From the SEM image of the broken surface of the faience bangle fragment, evidences of alterations were noticed before it was fired. The colour darkened (lightened in the SEM image) as we moved towards the interior of the bangle fragment (Figure 7 *b* and *c*). The variation in grain size could also be observed. The grain size became coarse as we moved towards the interior of the bangle fragment. The grains present in the fragment body were found to be randomly oriented, whereas those on the surface layer were tightly packed and homogeneous giving a finer look. It appeared as if the surface layer had been subjected to a surface whitening agent or an alkaline solution (perhaps $CaCO_3$). This was further confirmed by EDS results which showed the presence of a large amount of calcium and carbon on the surface, with their concentration decreasing towards the centre, suggesting that the sample was dipped in $CaCO_3$ or free lime solution before it was fired (Figure 7 *b* and *c*).

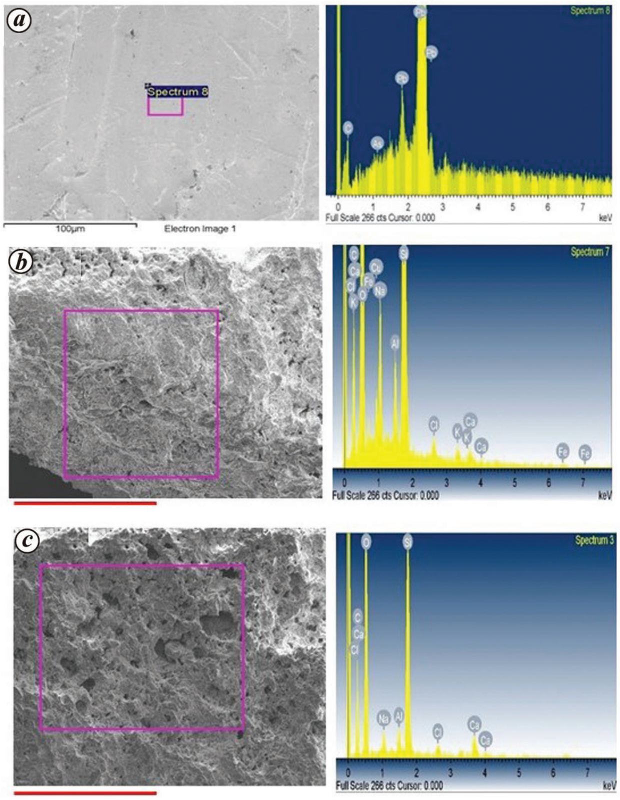

Figure 7. SEM images: (***a***) Lead nodule, (***b***) bangle fragment (from edge) and (***c***) bangle fragment (from centre) with EDS graph. Red bar is 800 µm.

Geological provenance to archaeological significance

This study aims to develop a mineralogical database of the major raw materials that were excavated from Dholavira. The mineralogical as well as chemical characteristics of these raw materials would be helpful to correlate with the adjoining sites or geological exposure, and thus are crucial for the interpretation of acquisition of these materials from different sites as well as from different geological exposures.

The raw material provenance has an indication from the major geological formations and lithotypes present in the adjacent regions of the Dholavira and Kachchh region as a whole. These regions (western India) have significant sources of minerals like quartz and its varieties like agate, jasper, carnelian, quartz, amethyst and opal. Agates are translucent microcrystalline silicates, whereas jaspers are opaque microcrystalline silicates having colours other than

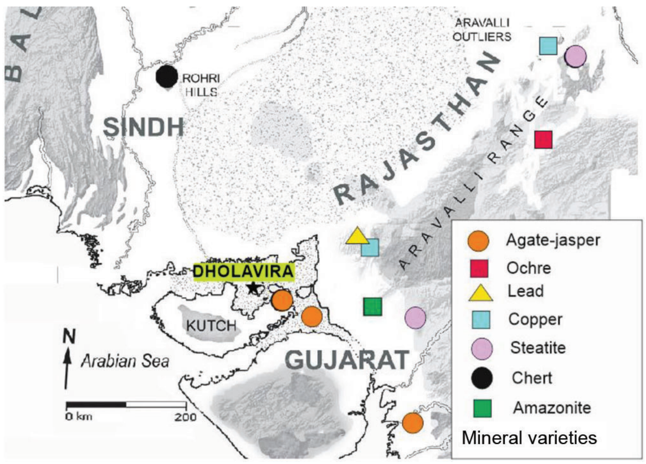

Figure 8. The potential geological sources of raw materials (minerals) possibly acquired by the Harappans.

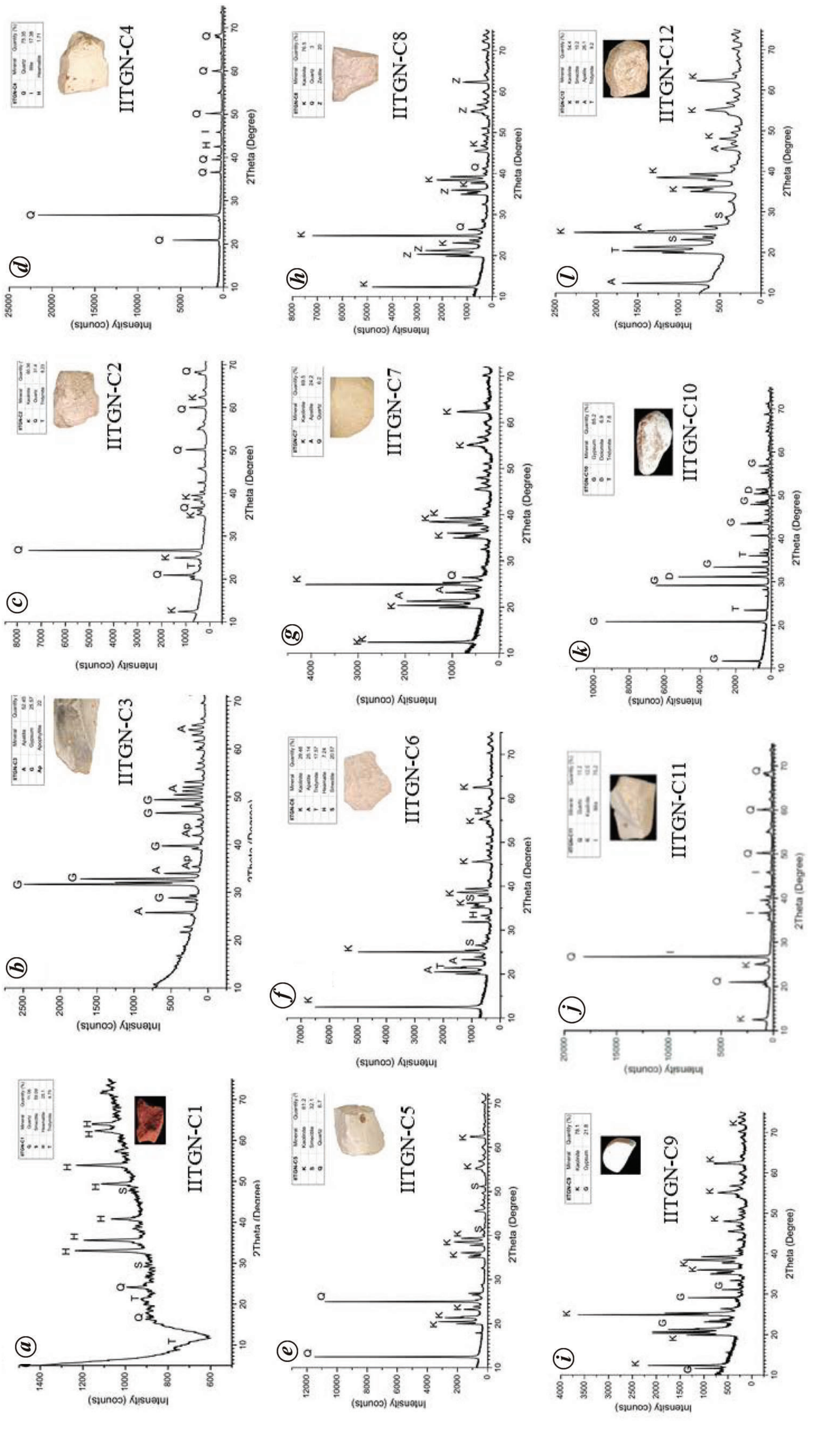

Figure 9. XRD pattern of selected clay samples from Dholavira: ***a***, IITGN-C1; ***b***, IITGN-C3; ***c***, IITGN-C2; ***d***, IITGN-C4; ***e***, IITGN-C5; ***f***, IITGN-C6; ***g***, IITGN-C7; ***h***, IITGN-C8; ***i***, IITGN-C9; ***j***, IITGN-C11; ***k***, IITGN-C10; ***l***, IITGN-C12.

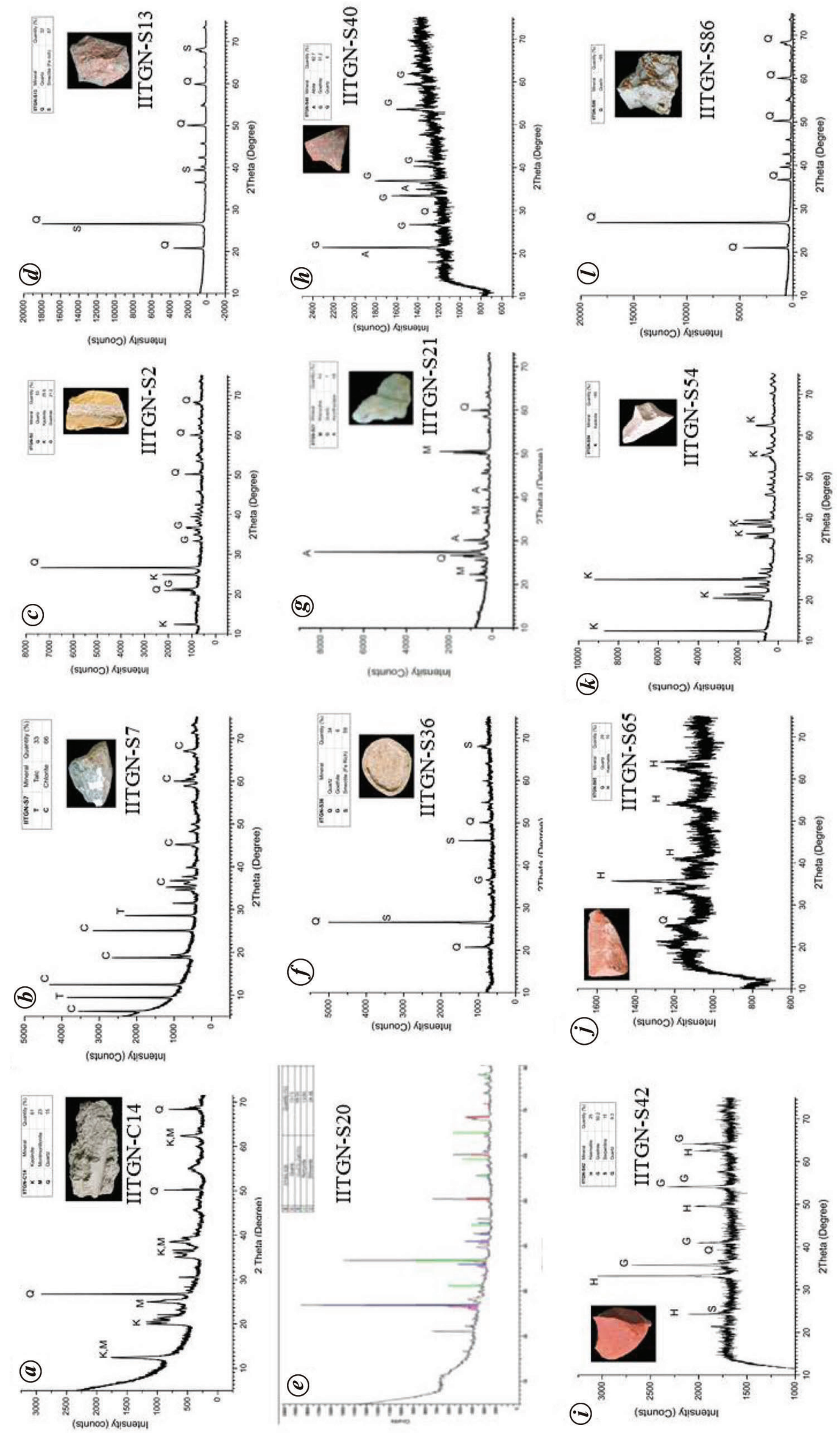

Figure 10. XRD pattern of selected lithic samples from Dholavira: ***a***, IITGN-C14; ***b***, IITGN-S7; ***c***, IITGN-S2; ***d***, IITGN-S13; ***e***, IITGN-20; ***f***, IITGN-S36; ***g***, IITGN-S21; ***h***, IITGN-S40; ***i***, IITGN-S42; ***j***, IITGN-S65; ***k***, IITGN-S54; ***l***, IITGN-S86.

chert colours (as defined in Law[6]). They were often developed as secondary minerals in the Deccan traps, either as fillings in the amygdaloidal cavities or as products of alteration and replacement. Amygdaloidal rocks are an excellent example of the primary geological context of agate. When these amygdaloidal rocks erode, loosened agates may fall away and be carried to a secondary geological context[6]. There is wide diversity and richness of agate exposures in Gujarat. The probable sources in Gujarat are Ratanpur, Central Kachchh, Khandek – eastern Kachchh, Mardek Bet – Little Rann, eastern Gujarat and Saurashtra[6]. There are a few sources of agate in Rajasthan, but these are not important due to the abundance of agate in close proximity to Dholavira. Chert, which forms due to sedimentary processes, has wide exposure in the Gujarat and Rajasthan region. For example, Rohri hill cherts are ubiquitous in many of the Harappan sites[14]. The quartz in clay samples indicates the fraction of sand and silt particles present in them, which are derived from alluvial and fluvial deposits of the Rann of Kachchh or Katol or Chari formations of Dholavira Island. The Katol and Chari formations of Kutch have a lithology of sandstone, shale and limestone. The significant kaolinite clay formed due to intensive chemical weathering of feldspar – mica-rich source rocks, e.g. granite, or arkosic sands is potentially exposed in the Khadir Island itself (at Jhandiyasar). Some of the clay samples revealed a significant amount of smectite (montmorillonite), which has high swelling and expansion properties, and were possibly used by the Harappans to construct reservoirs as plastering and mortar materials as they have adhesive and impermeable nature. Apatite was found in some clays, a good source of calcium and phosphorus. It may act as a component of a fertilizer and was possibly used by the Harappans in agricultural activities. During field visits, it was observed that the Khadir island has a large exposure of clay mixed haematite. Ochre-stone and other haematite-rich rocks are excellent examples of these deposits. They are mainly sedimentary deposits formed along river banks. Gypsum and limestone in clays are mostly the results of anhydride deposits in the riverbeds nearby. The Katol and Chari formations have several limestone deposits contributing significant amounts of limestone and calcium salts to the clay. Amazonite generally occurs in pegmatites (coarse-grained igneous rock). The probable source of the greenish variety of microcline is pegmatite from the Virpur granite area lying east and southeast of Palanpur[23]. Amazonite pebbles originating from the rocks can be found in the beds of the adjacent Sabarmati River[6]. The metallic raw materials like copper and lead nodules have wide exposure to ore in Rajasthan (Aravalli and Delhi Supergroup). The closest probable source of a lead nodule is the Ambaji area in northern Gujarat; a handful of lead samples from this source have been obtained previously[6]. With these basic interrelations of archaeological materials and their geological sources, a potential provenance map has been generated (Figure 8).

Conclusion

In the present study, clay and lithic raw materials were subjected to XRD and FE-SEM analysis. The lead nodule (95% Pb) was probably smelted or extracted Pb. SEM analysis of the faience bangle fragment indicated prior alteration when it was fired. The whitening agent used for the outer surface of the bangle was mostly $CaCO_3$. The ernestite nodules from the Sorath Harappan site of Bhagatrav were similar to samples from other Harappan sites in Gujarat. They had been used extensively for bead perforation even in the peripheral part of the main city site like Dholavira. The mineralogical interpretation of various microcrystalline varieties of quartz such as agate, jasper, carnelian, opal, chert, etc. was geologically derived probably from amygdaloidal Deccan Basalt that were abundant in Kachhch and Saurashtra. Further, cherts are having a vast exposure at Rohri hills. This could be a potential source of various chert tools found at the Harappan sites including Dholavira. Significant clay and haematite exposure was found at the Jhandiyasar site (north of Dholavira) at Khadir through field observations. Some metallic raw materials like Cu and Pb nodules were probably sourced geologically from the Aravalli and Delhi Supergroups from Rajasthan.

The raw material acquisition can be further validated by comparing it to the mineralogical database from other sites as well. SEM-EDS analysis is useful to understand the morphological structure, micro-grain size variability and elemental composition and its variation in varying spatial regions of the material. The present study indicates that the Harappans were skilful and well aware of the quality of raw materials used for different purposes.

Conflict of interest: The authors declare that there is no conflict of interest.

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