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Origin of the Harappan Ernestites: Geochemical Insights into Provenance and Fabrication

M. K. Mahala[1], Jyotiranjan S. Ray[1, *], A. K. Kanungo[2], G. N. S. Sree Bhuvan[3], A. Chatterjee[1,4], B. G. George[5], N. Sorcar[6,7], Y. S. Rawat[8], J. S. Kharakwal[9], and S. V. Rajesh[10]

[1]Physical Research Laboratory, Navrangpura, Ahmedabad 380009, India
[2]Indian Institute of Technology Gandhinagar, Palaj, Gandhinagar 382055, India
[3]Department of Earth Sciences, Pondicherry University, Puducherry 605014, India
[4]Department of Geology, Presidency University, College Street, Kolkata 700073, India
[5]Department of Earth Sciences, Indian Institute of Technology Bombay, Mumbai 400076, India
[6]National Centre for Earth Science Studies, Akkulam, Thiruvananthapuram 695011, India
[7]Korea Polar Research Institute, Incheon 21990, Republic of Korea
[8]Archaeological Survey of India, Dharohar Bhawan, 24 Tilak Marg, New Delhi 110011, India
[9]JRN Rajasthan Vidyapeeth, Sahitya Sansthan, Udaipur 313001, India
[10]Department of Archaeology, University of Kerala, Thiruvananthapuram 695581, India

*Correspondence: jsray@prl.res.in

Abstract

Advancements in stone bead technology, particularly in drilling techniques, emerged during the Indus Valley (Harappan) civilization. Long-constricted cylindrical drill bits, made from a unique stone called Ernestite, were a distinctive feature of this culture. The origin of Ernestite remains uncertain, as it has no known natural analogue and its synthesis process is poorly understood. This study presents a mineralogical and geochemical investigation of Ernestite stones and drill bits from multiple Harappan and contemporaneous sites in Gujarat, India, to uncover their origin. The $^{87}Sr/^{86}Sr$ and $\varepsilon_{Nd}(0)$ of the drill bits overlap with those of the Ernestite stones, confirming their genetic relationship. The texture and presence of pseudo-mullite ($SiO_2 > 40$wt%) with high Al-Ti-bearing hematite suggest that Ernestites are synthetic, created through a sintering process at ~1100°C. An abundance of sand to silt-sized detrital quartz, along with Fe-Ti-Zr-rich minerals, indicates the use of unevenly powdered sandstones and laterites as raw materials, with geochemical ties to regional sources.

Introduction

The prehistoric Indus Valley Civilization (IVC), also known as the Harappan civilization, was one of South Asia's most advanced civilizations of its time, renowned for its sophisticated urban architecture and material culture[1–3]. This civilization is famous for its fortified structures, efficient drainage systems, standardized seals and weights, and advanced technology employed in the manufacture of a diverse range of artifacts crafted from stone, metal, and shell[4–7]. Findings from nearly 2,500 sites across diverse geographic zones reveal that this civilization had broader spatial coverage compared to the contemporary Mesopotamian and Egyptian civilizations[5–8]. Most of the Harappan sites have been discovered along the river valleys of the Indus and Ghaggar-Hakra systems, distributed across Afghanistan, Pakistan, and northwestern India[9,10]. It is generally believed that the Harappan culture began as small agro-pastoral communities in its Early phase (>5000-2600 BCE), which matured into an urban civilization, recognized as the Harappan phase (2600-1900 BCE), demonstrating remarkable advancements in town planning, food production, and the technology of pottery and bead manufacturing. Subsequently, the society declined through de-urbanization in the Late Harappan phase (1900-1300 BCE)[10,11].

Stone beads are one of the critical indicators of cultural and trade practices within prehistoric South Asian civilizations[6,7]. The manufacture of stone beads began with the perforation of soft stones (e.g., limestone, steatite, and lapis lazuli) and later with hard stones (e.g., chert, agate, and jasper). The earliest evidence of stone beads dates back to the Mesolithic period (e.g., Jwalapuram)[12]; significant developments in bead production technologies, such as drilling, shaping, coloring, and mounting onto ornaments, occurred in the Neolithic and Chalcolithic periods[2] and became a key component of regional and external trades during the Harappan civilization[5,7]. Ancient Gujarat was well known for its rich agate resources, which attracted the Harappans to this region, and bead manufacturing industries/workshops were established in several urban centres in Kutch and Saurashtra[7,13]. Although various beads of different materials were in use, the long cylindrical beads of harder stones, typically jasper and carnelian, were manufactured through perforation using constricted cylindrical drill bits cut out from unique chips/stones called Ernestites[14,15], since their hardness is higher than agate (~7.5 on Mohs' scale[5,13,16]). The beads are characterized by a drill hole section with a stepped profile, as the drill bits are typically wide at the tip and narrow at the mid-section[7] (Fig. 2b).

The name "Ernestite" was given temporarily by Kenoyer and Vidale[14] after Ernst J.H. Mackay, but it remains in use. Ernestites are a signature finding of the urban phase of the Harappan civilization; however, they have been reported in large numbers from the late phase, single-cultured Harappan and Sorath Harappan sites as well[17,18]. Many Ernestite stones and drill bits have been found in close association with bead workshops in several Harappan sites in Pakistan (e.g., Harappa, Mahenjo-daro, Chahnudaro)[7,14] as well as in India (e.g., Dholavira, Khirsara, Kanmer)[15,17,19], and in a few Sorath Harappan sites such as Bhagatrav, Bagasra, Shikarpur, Nagwada[18,20–22]. Some important Harappan and Sorath Harappan sites, including those where Ernestites have been reported, are shown on the map (Fig. 1). Primarily manufactured by the artisans of the Harappan civilization, these unique materials almost became extinct in subsequent cultural periods[5].

Kenoyer and Vidale[14] described Ernestite at Mohenjo-daro as a rock composed of a mottled greyish-green to yellow-brown matrix with dark brown to black irregular patches or dendritic formations. Based on the XRD analysis of samples from Mohenjo-daro, Chanhudaro, and Harappa, they opined that these are metamorphic rocks composed of quartz, sillimanite, mullite, hematite, and titanium oxide phases. Law[7] observed significant quartz, mullite-sillimanite, and hematite phases in two samples from Harappa, as well as mullite and cristobalite in the other two. He found from XRD and EMPA analyses that the light and dark matrices consisted of clay-sized (<2µm) Al-Si bearing phases, compositionally similar to mullite and sillimanite, in addition to quartz. The dark matrix contained additional phases such as hematite, titanohematite, rutile, and zircon. He suggested that the Ernestite is likely a highly indurated tonstein flint clay, sufficiently heat-treated (up to 1100°C) to yield its characteristic hardness, based on the limited mineralogical and chemical data from his study and earlier experimental studies on clays. Tonstein is a kaolinitic (flint) claystone formed by diagenesis of volcanic ash in a swampy or non-marine environment[23]. Law[7] did suggest a few probable locations for the sources of tonstein closer to the city of Harappa; however, he did not provide experimental proof of the transformation of any natural rock or mineral to Ernestite by heating. His study attempted to explain the presence of Ernestite constituent minerals like zircon and rutile by attributing their likely occurrence in tonstein. Besides, no reasoning was provided for the observation that the mullites in the Ernestites had significantly different compositions than those that are stoichiometry mandated[24].

Because of the sheer number of Ernestite drill bits reported from the Harappan city of Dholavira in Gujarat (1212), Prabhakar et al.[15] hypothesized that the sources of Ernestite raw materials were located within the Kutch province of Gujarat. The XRD analyses of two samples of Ernestites from Dholavira and one sample from Bhagatrav, done by Prabhakar et al.[15] and Prasad and Prabhakar[17], respectively, showed the presence of quartz, hematite, and sillimanite/mullite. No cristobalite has been reported in Ernestites from any of the Indian sites. An ambiguity persists about the provenance (source regions) of the Ernestite raw materials. Earlier workers[7,15] speculated both local (Kutch/Ratanpur) and regional (Gujarat) sources, and there exists no isotopic data to establish the source(s) unambiguously.

Despite their ubiquitous presence in the Harappan settlements (Fig. 1), the origin of the Ernestite stones and drill bits remains uncertain. Hence, deciphering the Ernestite source materials and their geologic origin is vital to understanding the stone drilling technology and the inter-regional communication network during the Harappan period. In this study, we have addressed the following poorly understood aspects of the Ernestites with detailed petrography, mineralogy, mineral chemistry, geochemical, and isotopic investigations from three Harappan sites (Dholavira, Khirsara, Kanmer) and one Sorath Harappan site (Bhagatrav) in Gujarat, India: (1) What are the characteristics of the Ernestites? (2) What raw materials were used for their manufacturing, and (3) What were the geologic sources for these raw materials? In addition, we have attempted to shed some light on the manufacturing process of these stones.

Methods

Owing to our limited access to the Harappan artifacts, only six samples could be included in this study, consisting of three Ernestite stone samples (Fig. 2a) and three drill bits (Fig. 2b) from four sites (Khirsara, Kanmer, Dholavira, and Bhagatrav; Fig. 1) in Gujarat. The sample from Kanmer is associated with the mature Harappan phase. It comes from the collection of Kharakawal et al.[19]. The Bhagatrav sample is related to the Sorath Harappan phase[21] and comes from the collection of Kanungo[22]. A sample from Dholavira represents the Mature/Late Harappan phase[15]. The stratigraphic contexts of the samples can be found in the references given for each location. The Ernestite from Bhagatrav was subsampled into three parts. There were two drill bits from Kanmer (the first and third from the left in Fig. 2b) and one from Khirsara. Two laterite samples and two sandstone samples from the island of Khadir, on which Dholavira is located, were also studied. These rocks belong to the Paleogene and Jurassic periods, respectively. The sandstones belong to the Khadir Formation, which is equivalent to the Jhurio Formation, and the laterites are correlatable to either the Matanomadh Formation or the Naredi Formation of the Kutch mainland[25]. A brief description of the geology of Kutch and Khadir is provided in the Supplementary Information. Because of their size and rarity, the drill bits were analyzed only for Sr-Nd isotopic compositions, whereas the stones were powdered for mineralogical, geochemical, and isotopic analyses.

Petrographic studies were conducted on thin sections of all three Ernestite samples using transmitted and reflected light. Grain size analysis was done using the inbuilt software (Stream Basic) associated with the petrographic microscope (Olympus® BX-53). The mineralogical compositions of the Dholavira and Bhagatrav Ernestites whole stone powders were determined by X-ray diffraction (XRD) using a Bruker D2 Phaser diffractometer at the Physical Research Laboratory (PRL). Raman spectra for selected phases were obtained using a Renishaw® Invia Reflex Raman microscope with a spectrometer with a 514nm laser excitation wavelength at Pondicherry University. The laser beam, with 0.5 to 5% power, was focused on the sample spot through an X50 microscope objective lens with an exposure time between 60 and 120 seconds. The instrument was calibrated using the Raman silicon band of 520.7 cm^{-1}, with the help of an internal reference material Si(100), before the samples were analyzed. The raw spectra were preprocessed and subsequently corrected for background.

In-situ mineral chemical analyses for the Ernestite stones (thin sections) were conducted using a CAMECA SX-5 Tactics Electron Probe Microanalyzer (EPMA) at the National Centre for Earth Science Studies (NCESS)[26]. Analyses were performed at 20 kV accelerating voltage and 20 nA beam current with a beam diameter of 1μm. Analytical spots were chosen at the center of individual mineral grains that were devoid of any cracks and had uniform Z-contrast. Analyses for the aluminosilicate matrix and titanohematite phases were carried out on spots away from quartz boundaries. X-ray elemental maps were also generated for all three samples (Supplementary Information). The major element contents of Ernestites were determined by X-ray Fluorescence (XRF) spectroscopy using a Rigaku® Supermini200 instrument at PRL and the pressed pellet method[27]. Multiple international rock standards were used for calibration, and the reference material OU-6 from the International Association of Geoanalysts (IAG) was used for accuracy and precision checks. The major element contents of laterites and sandstones were measured at the NCESS, using an S4 Pioneer sequential wavelength dispersive-XRF[28], with reference materials VL-1 and MAG-1 used for accuracy and precision checks (Supplementary Data 1).

Bulk sample geochemical and isotopic measurements were carried out at PRL. About 50 mg of sample powder each was digested using conventional HF-HNO_3 and HF-HNO_3-HCl dissolution protocols for trace element and isotopic analyses, respectively. The details of the analytical procedures are given in George and Ray[29]. Trace element concentrations were measured on a Thermo® HR-ICPMS using BHVO-2 (USGS) as a calibration standard. Machine drift correction was performed using ^{115}In as an internal standard. The accuracy and precision of our measurements, determined by repeated analyses of BHVO-2 (as unknown), were better than 2% for REE and 5% for other trace elements. Sr and REE were separated from digested solutions by conventional cation exchange column chromatography using AG 50W-X8 resin (BioRad®), and Nd was eluted from REE using Ln-specific resin (Eichrom®), using protocols given in George and Ray[29]. Sr and Nd isotopic ratio measurements were performed on a TIMS (Thermo® Triton Plus) in static multicollection mode. Sr isotopes of some samples were measured on an MC-ICPMS at PRL[30]. Instrumental mass fractionation for Sr and Nd isotopic ratios was corrected using exponential fractionation (internal) correction equations of Thirlwall[31] and assuming $^{88}\mathrm{Sr}/^{86}\mathrm{Sr} = 8.375209$ and $^{146}\mathrm{Nd}/^{144}\mathrm{Nd} = 0.7219$. Multiple measurements of SRM-987 and JNdi-1 over three years yielded an average of $^{87}\mathrm{Sr}/^{86}\mathrm{Sr} = 0.710249 \pm 0.000009$ (2σ; n = 14) and $^{143}\mathrm{Nd}/^{144}\mathrm{Nd} = 0.512102 \pm 0.000010$ (2σ; n = 14).

Results

Petrography and Mineralogy

All Ernestite stone chips from Dholavira, Kanmer, and Bhagatrav exhibit heterogeneous physical appearances, unlike other Harappan artifacts, which demonstrate remarkable homogeneity[7] (Fig. 2a). They are hard (harder than quartz), highly compact, do not produce streaks, and are difficult to break. Two clear domains, a yellowish-brown or khaki color phase and a black color phase, can be distinguished by the naked eye (Fig. 2b). Transmitted and reflected light microscopy reveals that Ernestite stones contain detrital subangular to subrounded quartz grains (sand to silt-sized) and angular to sub-angular opaque phases like hematite and ilmenite set in a compact, fine-grained, light-colored (yellowish/khaki) groundmass of unidentifiable mineral(s) (Fig. 3). Quartz in Dholavira Ernestite occurs as fractured angular to subangular grains (Fig. 3a,b) compared to the sub-angular to sub-rounded grains in Bhagatrav (Fig. 3c,d) and Kanmer (Fig. 3e,f). The opaque phases (hematite, titanohematite, and ilmenite) appear as narrow bands or irregular patches. They occur in lower proportions in the Dholavira Ernestite than in the Kanmer and Bhagatrav stones. Hematite appears gray and displays the characteristic reddish internal reflection under plane and cross-polar view, respectively, in reflected light (Fig. 3g,h), and is often associated with ilmenite (shows bi-reflectance). All these detrital phases are essentially larger than clay-sized (~4μm) mineral grains that constitute a claystone. Sand-sized (210-736μm diameter; Supplementary Figure 1) detrital (clastic) grains of ilmenite and its partial replacement by hematite are also observed in the Kanmer Ernestite under a cross-polar view in reflected light (Fig. 3e,f). Zircon and rutile in Kanmer Ernestite have subrounded to irregular grain boundaries, confirming their detrital nature (Supplementary Figure 3). The size (longest diameter) distributions of detrital quartz grains (measured in the thin sections) in Dholavira, Kanmer, and Bhagatrav Ernestites are presented in a box plot (Fig. 4). Their ϕ (= $-\log_2 d$; d = diameter) sizes (1.84-6.64) vary between medium sand to fine silt, with half of the distributions falling between very-fine sand to coarse silt fractions. The quartz grains in Kanmer and Dholavira stones are moderately sorted ($1\sigma = 0.81$ and 0.71, respectively), whereas those in the Bhagatrav stone are moderately well-sorted ($1\sigma = 0.68$). Powder XRD patterns of the Dholavira and Bhagatrav samples (Supplementary Figure 2) reveal that quartz is the most abundant phase in all the samples, followed by a mullite-like phase (mullite/sillimanite). Hematite was detected only in the Bhagatrav dark matrix (Supplementary Figure 2), though it is observed in the petrography of all Ernestites.

The background-corrected Raman spectroscopic data for the Ernestite stone samples are given in Supplementary Data 3, and the spectra are presented in Fig. 5. Each trace in Fig. 5 represents a Raman (shift) spectrum for one spot analysis. Characteristic peaks for various minerals were identified by comparing our data with the reference spectra[32,33]. The laser excitation of quartz grains in Kanmer and Bhagatrav samples yielded good Raman signals (Fig. 5b-c). However, the excitation created a high fluorescence effect in the Dholavira sample, obscuring the Raman signal (Fig. 5a). We could observe the characteristic 468 cm^{-1} (sharp) peak of quartz only for four spots (Fig. 5b-c); whereas it was small and weaker in others. We did not observe the characteristic peaks for cristobalite (114; 231; 418 cm^{-1})[33]. Due to their intimate association with disseminated fine-grained quartz grains, the aluminosilicate matrix in the Dholavira and Kanmer samples did not yield discernible Raman spectra. However, the Bhagatrav sample yielded good spectra because of its discrete ellipsoidal occurrence as the dark matrix. From these spectra, we could identify the matrix mineral to be mullite (Fig. 5c-d). We did not find any indication of the presence of sillimanite (e.g., 235; 311; 872 cm^{-1})[34].

Major and Trace elements

The major oxide and trace element contents of two Ernestite samples from Bhagatrav and Dholavira, as well as two laterite and two sandstone samples from Khadir Island, are presented in Supplementary Data 1. SiO_2 content (47-61 wt%) is the highest among all oxides, with Al_2O_3, FeO^T, and TiO_2 being other major components. MnO, Na_2O, and P_2O_5 are either very low (< 0.1wt%) or absent, whereas K_2O and MgO concentrations are minor. Bhagatrav Ernestite has lower SiO_2 and Al_2O_3, FeO_T, and TiO_2 than Dholavira Ernestite. The major oxide data of the laterite and sandstone samples from the Khadir Island are also presented in Supplementary Data 1. Laterites have high Fe_2O_3 (36.7-37.6 wt%), moderate SiO_2 (32.31-32.61 wt%) and low Al_2O_3 (8.77-8.89 wt%), TiO_2 (1.32-1.34 wt%) contents, whereas sandstones are characterized by high SiO_2 (67.57-68.42 wt%), moderate Al_2O_3 (13.26-13.19), K_2O (1.83-1.84 wt%) and low Fe_2O_3 (2.06-2.09 wt%). Various oxides vs. SiO_2 diagrams plotted for Ernestites, sandstones, and laterites, along with the published data for Mesozoic sandstones[35,36] and laterites of Kutch[37–39], are presented in Fig. 6. Figure 7 presents the primitive mantle (PM) normalized multi-element patterns for the Ernestite samples and those for Mesozoic rocks[40], and laterites of Kutch region[39].

Mineral Chemistry

Representative backscattered electron (BSE) images of various phases in a polished thin section of the Kanmer Ernestite are given in Supplementary Fig. 3. Mineral compositions of different phases are provided in Supplementary Data 2. X-ray elemental maps for all three Ernestite stones (i.e., Kanmer, Bhagatrav, and Dholavira), as well as chemical spot analysis data (both by EPMA), are provided in Supplementary Figs. 3-6 and Supplementary Data 2. Quartz (SiO_2: 98-100 wt%) of varying sizes is dispersed within the light-colored (yellow) fine matrix, which is mainly composed of aluminosilicate phases (SiO_2: 40-53 wt%; Al_2O_3: 40-50 wt%). Although identified as mullites by XRD, the aluminosilicate matrix phases contain much higher SiO_2 than that mandated by stoichiometry (i.e., ≤ 30wt%)[24], therefore, we identify these phases as pseudomullites. Fe-Ti bearing phases, such as hematite (FeO: 71-74 wt%) and ilmenite (TiO_2: 51-56 wt%), often occur as narrow patches or are finely dispersed within the light (yellow) matrix. Many hematite grains have a high TiO_2 content (29-40 wt%) and can thus be classified as titanohematite. The titanohematites also contain an appreciable amount of Al_2O_3 (5-21 wt%).

Sr-Nd isotopic ratios

Results of Sr and Nd isotopic compositions of Ernestite stones and drill bits, laterites, and sandstones are provided in Table 1. The $^{87}Sr/^{86}Sr$ and $\varepsilon_{Nd}(0)$ of Ernestite stones and drill bits from Kutch (Dholavira, Kanmer, and Khirsara) vary between 0.71000 and 0.72282, and -14.7 and -13.9, respectively. In contrast, the Sr-Nd isotopic compositions of Bhagatrav Ernestites are more radiogenic in Sr and less radiogenic in Nd ($^{87}Sr/^{86}Sr$ = 0.73022 to 0.730876; $\varepsilon_{Nd}(0)$ = -18.3 to -18.1). The drill bits from Kanmer have an identical $\varepsilon_{Nd}(0)$ of -13.9, and one of the drill bits has almost identical $^{87}Sr/^{86}Sr$ as that of the Ernestite stone (0.72282 vs. 0.72207; Table 1). The laterite samples collected from the Khadir (Dholavira) have $^{87}Sr/^{86}Sr$ varying from 0.7089 to 0.7096, and their $\varepsilon_{Nd}(0)$ ranges from -7.7 to -7.5, whereas the sandstones have more radiogenic Sr and Nd ($^{87}Sr/^{86}Sr$ = 0.71494 and 0.74344; $\varepsilon_{Nd}(0)$ = -23.5 and -17.7).

Discussion

In the first-ever detailed characterization, Kenoyer and Vidale[14] suggested a metamorphic origin for Ernestites based on their identification of the matrix phases as sillimanite and mullite. Mullite is a rare mineral and has only been reported from specific contact-metamorphic rocks (in metamorphosed clays) and pseudotachylites[41,42]. It is also commonly observed in high-temperature ceramics and has been synthesized by heating various aluminosilicate minerals (e.g., kaolinite, kyanite, andalusite, sillimanite) at high temperatures (>1100°C)[43–45]. However, we identify these phases, which exhibit identical XRD and Raman spectra to mullite, as pseudomullites based on their higher SiO_2 contents (> 40wt%). Since mullite and pseudomullite are isostructural, all earlier studies, which relied primarily on XRD data, had incorrectly identified pseudomullite as mullite. "Pseudomullite" refers to a structure or phase that resembles mullite (Fig. 8) but is not the true, stoichiometric mullite. It is known to form during the thermal decomposition of kaolinite or other aluminosilicate materials[44]. Mullite refers to an experimentally observed solid solution series $Al_{4+2x}Si_{2-2x}O_{10-x}$ with $0.2 < x < 0.9$ (Fig. 8)[23]. According to Lenz et al.[24], the stoichiometric composition of synthetic mullite commonly varies between $3Al_2O_3.2SiO_2$ (~72wt% Al_2O_3) and $2Al_2O_3.SiO_2$ (~78wt% Al_2O_3). In natural mullites, Fe_2O_3 substitutes Al_2O_3, producing a wide range of compositions at ~30wt% SiO_2 (Fig. 8)[23,41]. In contrast, stoichiometric sillimanite has ~61 wt% Al_2O_3 (Fig. 8). The aluminosilicate phase in the Ernestite matrix is pseudomullite. It has higher SiO_2 and lower Al_2O_3 than those of natural mullite or sillimanite (Fig. 8). Contrary to an earlier report of the presence of sillimanite in the Ernestites[17], our Raman spectroscopic data indicate the complete absence of sillimanite in these stones (Fig. 5). Pseudomullites are not found in nature. However, it has been observed in synthetic materials produced by heating, such as ancient clinky potteries[46], that these materials likely developed at lower temperatures (≤1100°C) as a precursor to the formation of mullite from kaolinite (at ≥1200°C)[45]. Therefore, the presence of pseudomullites unambiguously rules out that Ernestites are natural rocks, indicating their origin by high-temperature processing. Absence of mullite also suggests that the sintering temperature never reached 1200°C.

The Harappans manufactured the Ernestites as the source stones for drill bits through a high-temperature heating process that could generate the pseudomullites. Further evidence for a high-temperature process is provided by the chemical composition of Fe-Ti-bearing phases, as determined by EPMA analyses. The presence of titanohematites with significant TiO_2 (29-40 wt%) and Al_2O_3 (5-21 wt%) suggests an extensive substitution between Fe_2O_3 and TiO_2 and between Fe_2O_3 and Al_2O_3. It is known that at temperatures below 800°C, only a limited solid solution between TiO_2 and Fe_2O_3 is possible[41]. Similarly, in the Fe_2O_3-Al_2O_3 system, higher Al_2O_3 (up to 10wt%) can be substituted into the hematite (Fe_2O_3) structure at high temperatures only (~1000°C)[41]. Therefore, higher TiO_2 and Al_2O_3 contents in the titanohematite confirm a heating process (>1000°C) in Ernestite manufacturing. It is thus apparent that the pseudomullite matrix was produced during high-temperature sintering. This provides the first geochemical evidence of sintering being used in the manufacture of Ernestites. This also successfully explains the presence of high-temperature craft objects, such as stoneware bangles[47,48], steatite beads[49], and furnaces[19], at the Harappan sites. The presence of detrital quartz grains, ilmenite, hematite, zircon, and rutile suggests that the raw materials used to make the Ernestites were natural.

Law[7] suggested tonstein as the only raw material for Harappan Ernestites. He attributed the coarser (up to 100µm) subhedral quartz or cristobalite grains (detected in his BSE images) to crystallization of free silica (released during heating) and zircon to a magmatic origin. He further proposed that the raw materials for the Ernsitites (i.e., tonsteins) were sourced from local/regional sources (i.e., Kutch). Tonsteins are hard and compact kaolinite-altered volcanic ash layers, generally found in coals and associated sediments[23]. These often contain magmatic quartz and zircon[23]. However, the microtextural characteristics of zircon, quartz, ilmenite, and rutile in Ernestites suggest their detrital (clastic) nature, making them unlikely to have been constituents of tonsteins. Additional evidence against using tonsteins for Ernestites comes from the presence of non-radiogenic Nd in the Ernestites ($\varepsilon_{Nd}(0) < -14$), because all volcanic ash beds in Kutch have been linked to the Deccan Traps[39], which contain more radiogenic Nd ($\varepsilon_{Nd}(0) > -11$, Fig. 9). Besides, our Ernestite samples contain sand-sized detrital quartz and ilmenite grains in contrast to a claystone/tonstein that usually contains clay-sized grains (≤ 2µm).

The size and moderately sorted nature of the detrital quartz grains suggest the use of coarser raw materials, such as sandstones, which were likely pounded into sand/silt-sized particles before being processed for sintering. It is possible that the Fe-Ti phases (hematite, titanohematite, ilmenite, and rutile) observed in the Ernestites also originated from the sandstones, as sandstones generally contain such heavy minerals. However, our Ernestites appear to exhibit mixing trends between the Mesozoic sandstones and laterites of the Kutch region in various oxide vs. SiO_2 plots (Fig. 6). Trace element patterns (Fig. 7) also suggest that such a mixture is necessary to explain the chemistry of the Ernestites. Besides, high contents of Al_2O_3 (> 20wt%) and high field strength elements (e.g., Sc, V, Cr, and Co) in Ernestites cannot be achieved by heating the sandstones of the Kutch alone. Therefore, a second end-member, containing Fe-Ti minerals but low in alkali elements, is needed to explain the Ernestite chemistry. The laterites of Kutch, derived from the mafic volcanic rocks of the Deccan Traps, fit the bill. The Paleocene to Eocene lateritic deposits in western Kutch (Matanomadh Formation) and Saurashtra (Jamnagar) contain both Al-rich (gibbsite, kaolinite) and Fe-rich (goethite, hematite, ilmenite-rich) phases and are depleted in alkalis[37–39], and have the required characteristics of this raw material. Although we discard claystone (tonstein) as the sole raw material, we do not deny its possible use in combination with sandstone and laterite for the Ernestite manufacturing.

Since kaolinite is a common mineral in laterites, it could have decomposed and undergone subsequent chemical and structural changes to form the pseudomullite matrix during the sintering process. The free (amorphous) silica released during the heating of pure kaolinite recrystallizes as cristobalite upon further heating to ~1350°C[50]. However, when kaolinite is heated with alumina-bearing material, no free silica forms[51,52]. Therefore, it is likely that during the sintering process carried out by the Harappans, free silica (SiO_2) formation was suppressed by the presence of gibbsite ($Al(OH)_3$) in the laterite. Moreover, gibbsite undergoes thermal decomposition to boehmite (AlO.OH) at 200°C, which transforms into a transitional alumina (α-Al_2O_3) phase at 500°C[53]. The Al_2O_3 in the titanohematite structure was likely sourced from gibbsite (lateritic) in the mixture during the α-Al_2O_3 stage. Furthermore, β-cristobalite has been observed at 1200-1300°C in quartz powder heating experiments[54]. The absence of this phase in the Ernestites, as confirmed by the Raman spectra of silica phases (Fig. 5), is consistent with our study's results, which indicate a maximum sintering temperature of 1100°C. We suspect that the cristobalites observed by Law[7] represent a higher temperature ($\geq$ 1200°C) or prolonged heating process.

The similar $^{87}Sr/^{86}Sr$ and $\varepsilon_{Nd}(0)$ values of Ernestite stone and drill bits from Kanmer genetically link the drill bits to the stone. Although it has been well established that Ernestite stones are the raw materials for long and constricted cylindrical drill bits[14], their isotopic similarity is the first-ever chemical evidence for the same. Because of the sheer number of Ernestite stones and drill bits from Dholavira, Law[7] speculated that the raw materials for the stones came from either the island itself (i.e., Khadir) or the Kutch region of Gujarat. However, our geochemical data (Figs. 6 and 7) support a regional sourcing of the raw materials. The sandstones and laterites of Kutch appear to have been the primary sources of the raw materials for the Ernestites. In search of more robust evidence for this geological provenance hypothesis, we make use of the Sr-Nd isotopic compositions of Ernestites and their potential source rocks (Table 1). The ε_{Nd} and $^{87}Sr/^{86}Sr$ of the Ernestites plot between the compositional ranges of the Mesozoic sandstones and the laterites of Kutch, and can be explained by a two-component mixing involving these two rock types. The isotopic compositions of laterites of Khadir, which are developed over volcanic ash depoits[39], fall well within the compositional field of the Deccan Basalts. This suggests that other lateritic horizons in the Kutch and Saurashtra region, developed over the Deccan Trap rocks, could also have served as sources for the raw material for Ernestites. A binary mixing model (Supplementary Figure 7) indicates a higher contribution (60-80%) from the sandstone end-member compared to the laterite end-member in most of our samples, with the contribution reaching as high as 90-95% for the Bhagatrav sample. Results of a Monte-Carlo simulation considering $\pm 2\sigma$ range for the above two end-member compositions (Fig. 9) strongly support the inference that the Harappans used laterite from different weathered (Deccan) horizons and sand from Mesozoic sandstones from Kutch to manufacture Ernestites.

We draw the following conclusions from our investigation of Ernestites, the parent material for the unique constricted drill bits of the Harappan Civilization, using petrographic, mineralogical, geochemical, and Sr-Nd isotopic techniques.

1. Stone drill bits have been isotopically fingerprinted to the Ernestites, confirming their genetic link.

2. The Ernestites consist of medium sand to fine silt detrital quartz, hematite, ilmenite, zircon, and rutile welded together in a fine-grained aluminosilicate matrix/groundmass.
3. Ernestites' texture (larger mineral grains and their detrital nature), and its Nd isotopic composition ($\varepsilon_{Nd}(0) > -11$) rule out the use of tonstein flint as a raw material.
4. The aluminosilicate matrix/groundmass phase has been chemically identified as pseudomullite, though its XRD and Raman spectra are similar to those of mullite.
5. The presence of pseudomullites, with high SiO_2 contents (> 40wt%), unambiguously makes Ernestites artificial, with supporting evidence from the significant substitution of Al_2O_3 and TiO_2 in hematites. These data also suggest a high temperature (reaching 1100°C) synthesis of Ernestites.
6. Mineralogy, texture, and mineral chemistry suggest Ernestites were manufactured through a high-temperature sintering process involving sand and clay-bearing raw materials.
7. Major and trace elements and Sr-Nd isotopic data point to the likelihood of the raw materials' regional provenance (sandstones and laterites of Kutch).
8. All our findings suggest that Ernestites were likely made in the Harappan centres of Gujarat, India, and the Ernestite-based drilling technology was exclusive to this civilization.

Author contributions

JSR and AC conceived the study. AKK, YSR, JSK, and SVR supplied samples. AC, MKM, BGG, NS, and GNSSB conducted the analytical work. MKM, GNSSB, AC, and JSR interpreted the data. JSR secured the project's funding, and all authors contributed to the writing.

Data Availability Statement

All data generated for this study are in the tables in the manuscript and the supplementary files.

Competing interests

The authors declare no competing interests related to this work.

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Appendix A. Supplementary Information

Supplementary materials for this article have been attached as separate files.

1) Supplementary Information (Doc file: Text and Supplementary Figures 1-7)
2) Supplementary Data 1 (Excel file: Major and Trace element data)
3) Supplementary Data 2 (Excel file: EPMA mineral chemistry data)

4) Supplementary Data 3 (Excel file: Raman spectroscopy data)

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Table 1. Sr-Nd isotopic data for the Ernestite stones/drills and Laterites

Sample	**Type**	**Location**	**^{87}Sr/^{86}Sr**	**^{143}Nd/^{144}Nd**	**$\varepsilon_{Nd}(0)$**
E-2	Ernestite	Bhagatrav	0.73022*	0.511708	-18.1
E-3	Ernestite	Bhagatrav	0.73027 *	0.511703	-18.2
E-4	Ernestite	Bhagatrav	0.73087*	0.511700	-18.3
3304	Drill bit	Kanmer	0.72282*	0.511927	-13.9
3285	Drill bit	Kanmer	0.71778 *	0.511926	-13.9
ERN-KM	Ernestite	Kanmer	0.72207*	0.511884	-14.7
ERN-DV	Ernestite	Dholavira	0.712901	0.511900	-14.4
ERN-KU	Drill bit	Khirsara	0.709991	0.511915	-14.1
KH-3	Laterite	Khadir	0.708990	0.512256	-7.5
KH-4	Laterite	Khadir	0.709551	0.512245	-7.7
KH-15-6	Sandstone	Khadir	0.714940*	0.511731*	-17.7
KH-15-27	Sandstone	Khadir	0.743439*	0.511433*	-23.5

Note: All ratios are TIMS data except those marked with *, which are MC-ICPMS data. The average isotopic ratios and external reproducibilities determined for the international SRM-987 and JNdi-1 in TIMS, after repeated analyses over three years, are ^{87}Sr/^{86}Sr = 0.710249±0.000009 (2σ; n=14) and ^{143}Nd/^{144}Nd = 0.512102±0.000010 (2σ; n = 14), respectively. $\varepsilon_{Nd}(0) = [(^{143}\mathrm{Nd}/^{144}\mathrm{Nd})_{sample}/^{143}\mathrm{Nd}/^{144}\mathrm{Nd})_{CHUR} - 1]\times 10^4$, where CHUR = Chondrite Uniform Reservoir and (0) stands for present-day value.

Figure Captions

Fig. 1. A schematic geographical map of western India and Pakistan showing important Harappan urban centers and cities from which Ernestite stones and drill bits have been reported. The three Harappan sites and one Sorath Harappan site, from which the Ernestite samples for this study came, are marked.

Fig. 2. The photographs of (a) Ernestite stones from Dholavira, Bhagatrav, and Kanmer, and (b) Ernestite drill bits from Kanmer studied in this work. Note the textural variations between different samples, as reflected in their colors. The first and third drill bits shown in (b) have been used for isotopic analyses.

Fig. 3. Photomicrographs of Ernestite thin sections: (a) sample from Dholavira in plane-polarized transmitted light showing the presence of quartz (Qz) and opaques (Opq); (b) same as in (a) with hematite (Hem) displaying characteristic red internal reflection under reflected lights; (c-d) sample from Bhagatrav in plane-polarized and cross-polarized transmitted lights; (e-f)

sample from Kanmer showing detrital ilmenite (Ilm) and quartz (Qz) in plane-polarized transmitted light; (g-h) sand-sized ilmenite (Ilm) grains and hematite (Hem) patches in the same sample as in (e) in plane and crossed polarized reflected lights, respectively.

Fig. 4. Boxplot showing grain size distribution (in ϕ scale) in the Ernestites. Relevant statistical information is given in boxes inside the figure. The mean (circle) and median (dashed line) are marked in each box. Bhagatrav-1 and Bhagatrav-2 represent the yellow (khaki) and black colored groundmasses, respectively. Symbols: n = number of observations; μ = mean; σ = standard deviation; SK = skewness; K = kurtosis.

Fig. 5. Raman spectra of silica and aluminosilicate phases in the Ernestite samples from (a) Dholavira; (b) Kanmer, and (c-d) Bhagatrav, identified as alpha-quartz and mullite, respectively, based on their Raman shifts. One sample (Bhagatrav-2) was also analyzed for its Raman shift between 1000 and 1500 cm^{-1}.

Fig. 6. Plots of various oxides vs SiO_2 for Ernestite samples, laterites, and sandstones from Khadir Island (data in Table 1). Compositions of Mesozoic sandstones[35,36] and laterites of Kutch[37–39] are plotted as fields for comparison.

Fig. 7. Primitive Mantle normalized spider diagram for Ernestite samples. Also plotted are the data for laterites[37–39] and Mesozoic shales[40] of Kutch. Normalizing values are from McDonough and Sun[55].

Fig. 8. The plot of Al_2O_3 vs. SiO_2 for the aluminosilicate phases in our Ernestite samples compared with compositions of natural mullite[24], stoichiometric (synthetic) mullite, mullite solid solution, and stoichiometric sillimanite.

Fig. 9. The plot of $\varepsilon_{Nd}(0)$ vs. $^{87}Sr/^{86}Sr$ of the Ernestite stones and drill bits, along with laterites and Mesozoic sandstones[56] of Kutch. The compositional field of Deccan Basalts[57] is shown as a pale cyan rectangle for comparison. The points represent the results of a Monte-Carlo binary mixing simulation involving the Mesozoic sandstone (average composition: $^{87}Sr/^{86}Sr$ = 0.733600±0.030 & ε_{Nd} = -18.7±6.2) and the laterite (average composition: $^{87}Sr/^{86}Sr$ = 0.709271±0.0008 & ε_{Nd} = -7.6±0.30), at 2σ levels of uncertainty, of the Kutch region. f_{ms} represents the fraction of sandstone end-member in the mixture.