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An Insight into Gandharan Art: Materials and Techniques of Polychrome Decoration

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Abstract: Gandharan art developed in the Himalayan area in the early centuries CE. It has been investigated mostly from an iconographic point of view, missing, until very recently, a systematic technical investigation of materials and techniques. Recently our team began performing chemical analyses of the traces of the polychromy originally covering statues, reliefs and architectural decorations, to discover the ancient painting techniques and artistic technologies. This paper presents the results of the analytical investigation (optical microscopy, Raman spectroscopy and gas chromatography coupled with mass spectrometry) of pigments, ground layers and binders of a new group of samples taken from stucco architectural decorations (2nd–3rd/4th centuries CE). The samples were collected directly at an archaeological site in the Swat Valley, ensuring the exact knowledge of their stratigraphic provenance, as well as the absence of any restoration treatment applied prior sampling. The results are discussed in the wider context of Gandharan polychromy investigated so far by our team, as found in sculptures and architectural decorations preserved in museums (in Italy and France) and in archaeological excavations in Pakistan. The aim of this research is to shed light on the materials and techniques of this Buddhist ancient art from this region and on the influences exerted on it from Eastern and Western artistic traditions.

Keywords: statues; reliefs; architectural decorations; stucco; pigments; supports; binders

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1. Introduction

Gandharan sculpture has been studied primarily from an iconographic point of view (see for example [1–8]). Research shows that this Buddhist ancient art, which developed in the Himalayan area (modern-day Pakistan and Afghanistan) in the early centuries CE, was influenced by Greco-Roman, Iranian, Indian and Central Asian artistic cultures (see for example [1,9–11]). Originally, these sculptures were very different in appearance compared to what is observable today.

Figure 1 shows some examples of sculptures and architectural decorations from the collection of the Musée Guimet, Paris, presenting traces of residual paint or preparation layers.

Figure 1. Paris, Musée Guimet: (**a**) polychrome Stucco Head of Salabhanjika from Hadda (Afghanistan) 3rd century CE (inv.MG 17203). (**b**) polychrome stucco relief from Hadda (particular), monastery of Tapa-i-Kafariha (Afghanistan), 2nd–3rd century CE (Barthoux expedition, 1928-unnumbered); (**c**) polychrome clay statue of seated Buddha, from the monastery of Fondukistan, Chorband Valley (Afghanistan), 7th century CE (inv. MG 18970); (**d**) polychrome clay statue of Two Nāga King (particular from the head) from the monastery of Fondukistan, Chorband Valley (Afghanistan), 7th century CE (inv. MG 18597); (**e**) polychrome schist relief from monastery of Shotorak (Afghanistan), 2nd–3rd century CE (inv. MG 22148). (© ICR, photo Simona Pannuzi).

The various colours present on the different parts of the body of the figures probably also had a religious meaning, as in Indian art [12,13]. Nowadays, just a few traces of polychromy can be found on Gandharan statues, reliefs and buildings. Although study is needed to reconstruct the original appearance of these artworks and buildings, only recently has this polychromy begun to be studied in depth. Moreover, following excavation, the early identification of polychrome and gilded traces is absolutely necessary to ensure the correct approach towards interventional conservation is followed, particularly to avoid aggressive cleaning steps that may erase and contaminate the few remaining traces that are left of the original decorations.

In the last few years, a programme of collaborative research has been established by the Italian Institute for Restoration (ICR), with the University of Pisa, the Italian Archaeological Mission in Pakistan (ISMEO-ACT Field School; ISMEO/Ca' Foscari University of Venice), the Stuttgart University, the Musée Guimet in Paris, the ex-National Museum of Oriental Art "Giuseppe Tucci" of Rome (ex MNAO, now merged into *Museo delle Civiltà*), the Oriental Art Museum of Turin (MAO) and the Civic Archaeological Museum of Milan. Research focuses on revealing the original appearance of Gandharan sculptures, reliefs and architectural decorations, disclosing the materials and techniques of Buddhist ancient art in this region, highlighting the influences exerted on it from Eastern and Western artistic traditions, and guiding conservation interventions. To this end, technical investigations have been carried out on the petrographic composition of the stucco architectural decorations found in the Swat Valley and petrographic and mineralogical composition of schist stone reliefs conserved in ex-MNAO [14,15], as well as on the painting technology on Gandharan stone, stucco and clay sculptures and stucco architectural decorations [16–19]. The state of conservation of the materials and polychromy has also been investigated in previous studies [20–22].

In this paper, we present new data on the use of pigments and binders on Gandharan stucco architectural decorations of religious buildings in the Swat Valley. The samples presented here add important information which is currently missing in the technical literature dealing with Gandharan art. Samples were collected directly from Swat archaeological sites (Figure 2), ensuring a clear knowledge on their stratigraphic provenance and the absence of any prior conservation or restoration treatment. The stucco samples examined in this study were all taken in the spring campaign of 2012, as part of the ACT-Field School project ("Archaeology-Community-Tourism"; 2011–2016; Pakistan-Italian Debt Swap Agreement). The sampling sites are located in the middle section of the Swat Valley (Khyber-Pakhtunkhwa province) in northern Pakistan. The samples were all exported with federal (Department of Archaeology and Museums, Govt of Pakistan) and provincial (Directorate of Archaeology and Museums, Govt of Khyber-Pakhtunkhwa) permission. The samples are representative of three sites, each of which is characteristic of a single Buddhist cult monument, and all three relating to the same chronological phase, i.e., the period between the first half of the 2nd century and the end of the 3rd century CE [23]. The analytical results obtained from these stucco samples were then compared to previously published samples taken from stucco architectural decorations and polychrome stone, stucco and clay sculptures and reliefs [14,16,17], to discuss what we know about the materials and techniques used by Gandharan artists.

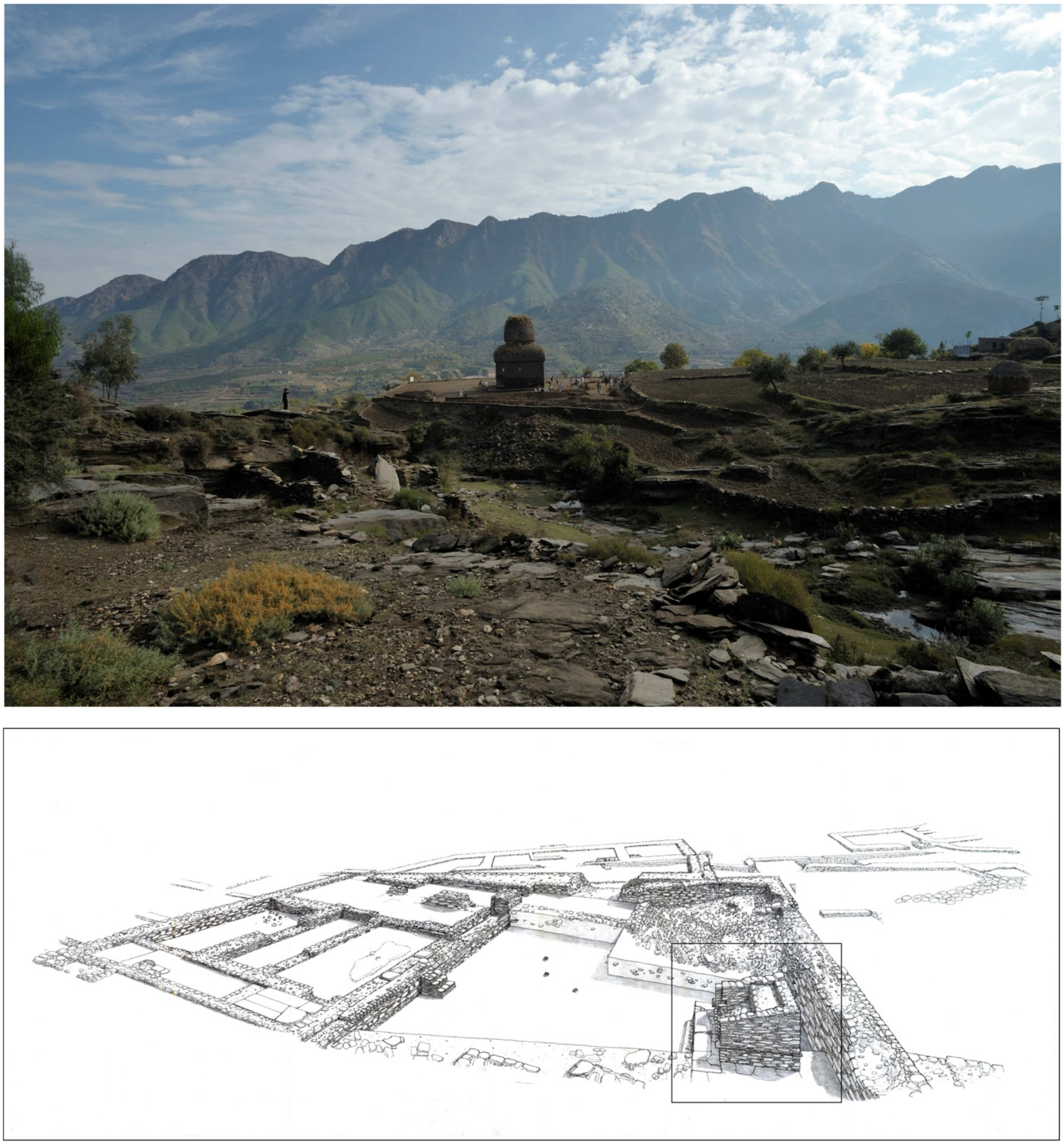

Figure 2. (**Top**): Gumbat/Balo Kale Trench I (seen from W) (© ISMEO/ACT; Photo by Edoardo Loliva); (**Bottom**): View from ENE of the precinct of Temple K: Shrine [1123–1023] is marked (© ISMEO/ACT; Drawings by Francesco Martore).

2. Materials and Methods

2.1. Samples

The samples analysed in this study came from stucco architectural decorations of buildings from three different locations: samples B8 and BKG 1123 (15) come from: Barikot, a fortified agricultural colony in existence between the middle of the 1st millennium BCE and the 4th century [24,25]; samples GBK (18), GBK (17) and A3 came from Gumbat/Balo Kale, an important sacred area about 5 km SW of Barikot [23,26]; and sample C11 came from the sacred area of Amluk-dara, about 5 km SE of Barikot [23]. Table 1 summarises the main features of the samples and the analyses performed on each of them as part of the present study.

Table 1. Description of the samples on stucco supports collected from architectural decorations analysed as part of the current study.

Sample Description					Analyses Carried Out		
Sample Name	**Colour of the Polychrome Layer**	**Presence of a Preparation Layer**	**Archaeological Site**	**Period**	**Raman**	**GC-MS**	**Sample Type**
GBK (18)	red	yes	Gumbat/Balo Kale (Pakistan)	end 2nd century–early 3rd century CE		x	powder
GBK (17)	red	yes				x	powder
BKG 1123 (15)	red	yes	Barikot (Pakistan)	middle-end 2nd century CE	x	x	fragment two sub-samples analysed (pl and prep)
C11	red	yes	Amlukdara (Pakistan)	late 3rd–early 4th century CE	x	x	paint layer
A3	black and blue paint layer	yes	Gumbat/Balo Kale (Pakistan)	beginning of the 2nd century CE	x	x	paint layer
B8	red	no	Barikot (Pakistan)	ca. 3rd century CE	x	x	paint layer

A detailed description of the samples from an archaeological point of view is provided in Supplementary Materials Section S1.

Samples (BKG 1123 (15), A3, C11 showed a white preparation layer (layer 1) between the coloured paint layer (layer 2) and the stucco support (layer 0). Sample BKG 1123 (15) consisted of a big fragment of around 4 cm in length, showing a red coloured area (layer 2) over a white preparation layer (layer 1). Two different subsamples were taken from BKG 1123 (15), one from the red paint layer, named BKG 1123 (15) _pl, and one from the white preparation below, named BKG 1123 (15) _prep. Sample A3 showed two different paint layers: a blue paint layer (layer 2A) applied onto the preparation layer (layer 1) and a black paint layer on top (layer 2B). Samples GBK (18) and GBK (17) instead were already powdered, and therefore, no separation between layers was possible prior to analysis. However, white grains from the preparation layer were observed under the microscope. Sample B8 showed no white preparation layer, and the red paint layer seems to have been directly applied onto the stucco support. The macrophotographs of samples C11, A3 and B8 are reported in Figure 3, together with a scheme reconstructing their respective stratigraphies. Samples GBK (18), GBK (17), and BKG 1123 (15) are shown in Supplementary Materials Section S1, Figures S1–S3.

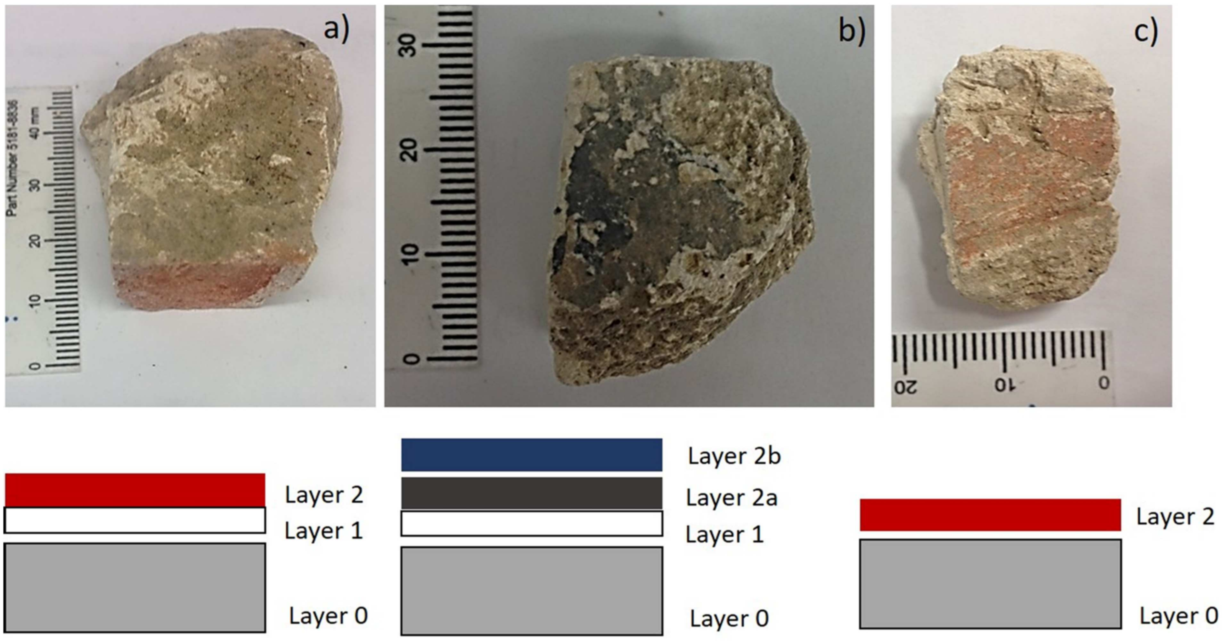

Figure 3. Macrophotographs of samples (**a**) C11; (**b**) A3; (**c**) B8 (**top**) and scheme of the respective stragraphies (**bottom**).

The stucco used as the support was most likely based on limestone, as other stucco samples from the same sites and chronologies were analysed by petrographic analysis (SEM EDS, XRD) in a previous study [14,17]. The limestone was not a local rock in Swat, but most likely originated from extended outcrops located in the relatively close northwest of Islamabad [14]. This stucco is composed of a mixture of fine-grained binder calcite matrix and local crushed rocks (i.e., schists and granites).

2.2. Instruments and Methods

2.2.1. Optical Microscopy

Micrographs were collected using an optical stereo-microscope (Leica M125). The sample build-up was observed and photographed using an optical microscope Leica DM-RXP, and when possible subsamples were collected using a scalpel.

2.2.2. Raman Spectroscopy

Raman analyses were performed directly on the polychrome surface of the samples. Spectra were recorded on a Raman Invia system (Renishaw) equipped with CCD detector with a resolution of 1800 lines/mm, coupled to an imaging microscope with 10×, 50× and 100×, magnifications. An HeNe laser at 633nm and a NdYag laser at 532 nm were used as excitation and were filtered to give a laser density power at the exit of the objective lens which varied up to a maximum of 2 W/mm^2. Several measurements were performed, adjusting the laser fluence to 0.5 W/mm^2, to ensure that the heating produced by the laser was minimised and the sample was not altered. Typically, a 50× magnification was used and the spot size diameter was about 5 µm. Raman spectra were collected from ten different points randomly from each sample under the same conditions. Each spectrum was averaged over four scans, corresponding to a total acquisition time of 30 s.

2.2.3. Gas Chromatography Mass Spectrometry (GC-MS)

Samples (5–500 mg) were subject to a multistep analytical procedure aimed at analysing lipids, proteins and saccharide materials in the same sample [27]. This is based on a set of extraction and hydrolysis steps, followed by silylation and GC-MS analyses, leading to three different chromatographic profiles, one for lipid derivatives, such as fatty and dicarboxylic acids, one for saccharides and one for amino acids. Analyses were carried out with a 6890N GC System Gas Chromatograph (Agilent Technologies, Palo Alto, CA, USA), coupled with a 5975 Mass Selective Detector (Agilent Technologies, Palo Alto, CA, USA) single quadrupole mass spectrometer, equipped with a PTV injector. Details of the analytical procedure and instrumental apparatus are reported elsewhere [27].

The identification of lipids, proteins and saccharide materials is based on the evaluation of the chromatographic profiles obtained and the quantitative analysis of monoacarboxylic acids, dicarboxylic acids, amino acids and aldoses and uronic acids (Supplementary Materials Section S2). Quantitative data are thus evaluated in relation to the blank of the procedure in order to determine whether analytes are present above or below the detection and quantitation limits of the procedure (Supplementary Materials Section S3). When analytes are below the detection limit, we can only conclude that the relative material, if present, is not detected. If saccharides are above the detection limit, the evaluation of the chromatographic profile may be carried out to determine the source of the saccharide material by carrying out a comparisons with a reference database [28]. If amino acids are above the quantitation limit, the quantitative profile of the sample is compared to a dataset of reference samples by means of principal component analysis in order to identify the source of the material [29]. When amino acids are above the detection limit, but below the quantitation limit, the identification of the source of the proteinaceous material cannot be carried out, as this is based on quantitative analyses. As hydroxyproline is a maker for collagen, the presence of this protein in the sample can be assessed based on the detection of this amino acid above the detection limit [27].

3. Results

The results obtained for the characterisation of the samples analysed are summarised in Table 2.

Table 2. Summary of the results on the organic and inorganic materials obtained from the analysis of the samples by Raman spectroscopy and GC-MS analyses. n.p. not performed; n.d. non detected: below limit of detection; LOQ: limit of quantitation.

Sample	Inorganic Composition		Binder Analysis			
	Raman Shift (cm^{-1})	Pigments Identified	Subsample Analysed	Lipid-Resinous Fraction	Saccharide Fraction	Proteinaceous Fraction
GBK (18)	n.p.		Paint and preparation layer powder (0.3 mg)	animal fat	n.d.	n.d.
GBK (17)	n.p.		Paint and preparation layer powder (0.7 mg)	animal fat	n.d.	n.d.
BKG 1123 (15)_pl	n.p.		Red paint layer	n.d.	n.d.	present, below LOQ
BKG 1123 (15)_prep	n.p.		White preparation layer	n.d.	n.d.	n.d.
C11	220, 290, 400, 605, 660 1086	Red ochre Calcite	Red paint layer	n.d.	n.d.	egg, animal glue (minor component)
A3	283, 1086 122, 195, 460 1335, 1590	Calcite quartz Carbon black (graphite)	Black and blue paint layer	n.d.	n.d.	n.d.
B8	220, 290, 395, 605, 660	Red ochre	Red paint layer	n.d.	n.d.	egg, animal glue (minor component)

3.1. Inorganic Composition

The Raman spectra in the range 200–2000 cm^{-1} of all samples analysed are shown in Figure 4.

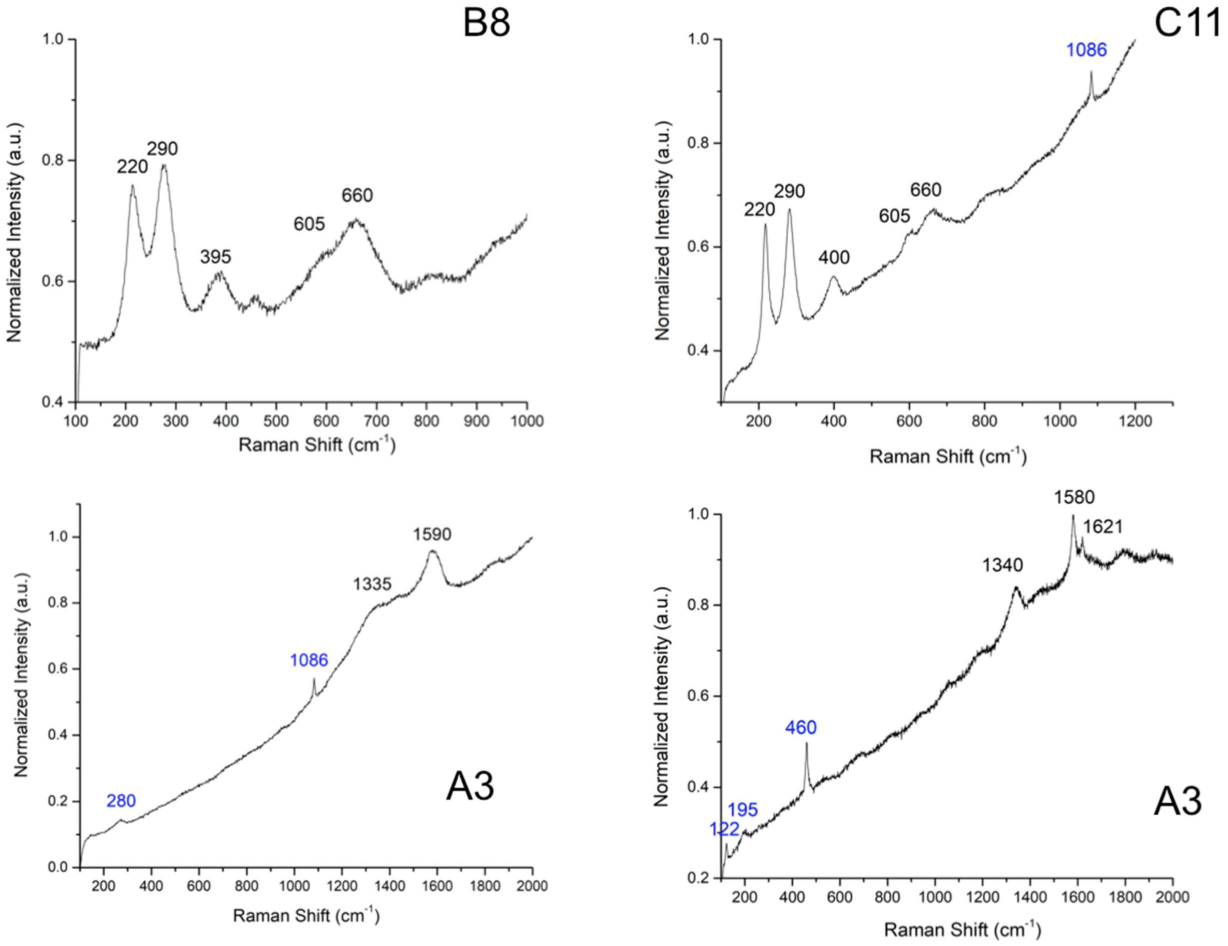

Figure 4. Raman spectra of samples B8, C11 and A3 (two different areas).

Calcite (signals at 280 and 1086 cm^{-1}) and quartz (signals at 122, 195 and 460 cm^{-1}) were identified [30] in samples A3 and C11, in agreement with the presence of a preparation layer in these samples, whereas their characteristic bands were completely absent in all spectra of sample B8.

The black decoration in sample A3 showed a typical signal of a microcrystalline graphite [31,32], as observed in the literature for an array of black-based pigments [33]. The blue pigment was not identified in this sample.

Red paint layers both in samples C11 and B8 showed the characteristic signals at 220, 290, 39, 605 and 660 cm^{-1} ascribable to hematite, suggesting red ochre [34].

3.2. Organic Composition

Fatty acids were detected below the detection limit of the procedure, and dicarboxylic acids were not detected, indicating the absence of a siccative oil. The only exceptions are samples GBK (18) and GBK (17), whose lipid-resinous fraction showed the presence of fatty acids, including those with an odd number of carbons: pentadecanoic acid (C15), heptadecanoic (C17), nonadecanoic (C19) and their isomers above the quantitation limit of the procedure. Their presence may be related to the occurrence of a fat of animal origin [35]. Saccharides detected were below the detection limit. We can thus conclude that, if saccharide materials were used as binders, these were highly degraded, and thus not detectable.

In Samples GBK (18) and GBK (17) amino acids were below the detection limit. Sample BKG 1123 (15) _pl showed the presence of a proteinaceous material above the detection limit of the procedure, but below the quantitation limit, preventing quantitative analysis of the profile to identify the source of the material. However, we may exclude the presence of animal glue based on the absence of hydroxyproline from the amino acid profile.

Samples C11 and B8 showed the presence of a proteinaceous material above the quantitation limit of the procedure. The relative amino acid percentage content (Supplementary Materials Section S4, Table S3) and its subsequent multivariate statistical treatment together with a data-set [29] of 121 reference samples of animal glue, egg and casein, using the PCA (Principal Component Analysis) method, shows that the samples are located in or very close to the egg cluster (Figure 5). Moreover, both samples also showed the presence of hydroxyproline, indicating that small amounts of animal glue are also present [27].

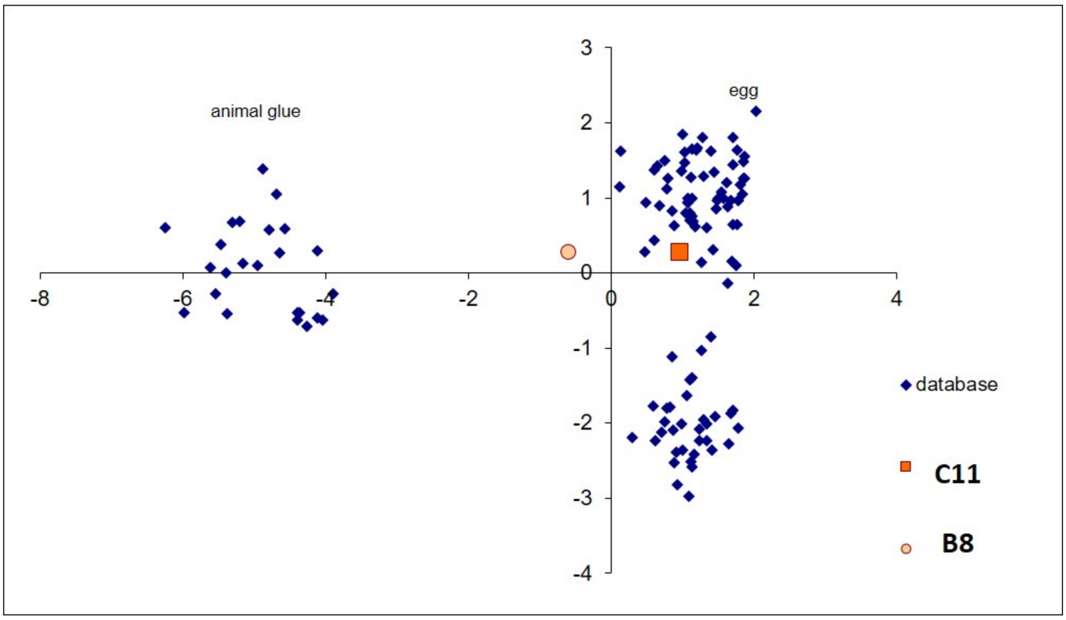

Figure 5. PCA score plot of the samples showing a protein content above the quantitation level.

4. Discussion in the Framework of Gandharan Art Technical Literature

In order to discuss the know-how of, and materials and painting techniques used by Gandharan craftsmen, the samples studied in this paper were compared with other samples already presented in previous studies [14,16–19].

Different types of objects have been studied, comprising different supports (stucco, clay or stone), provenance (archaeological context or museum collection) and type (decorative architectural decorations or statues). FTIR, SEM-EDS, Raman and XRD were used to characterise the support, preparation layers and identify the pigments used in the polychrome paint layers. Of these, 19 samples were studied by GC-MS and proteomics in order to establish the presence of an organic binder, determine its nature and, possibly, identify its origin. A summary of previously published data is reported in Supplementary Materials Section S4.

4.1. Gandharan Polychromy: Support

Three main supports may be distinguished in Gandharan polychromy: stone, clay and stucco.

4.1.1. Stone

Stone supports of statues and reliefs have been analysed previously on a wide number of samples coming from artworks conserved in ex-MNAO of Rome. Serpentinite, talc schist and serpentine schist were identified [15].

4.1.2. Clay

Two clay samples taken from artefacts (probably sculptures) found in archaeological sites in Afghanistan were analysed by means of petrographic microscopic analysis, scanning electron microscopy, and X-ray diffraction: Tapa Sardar and Tape Narenj [14]. The sample clay artefact from Tapa Sardar showed the presence of feldspar, quartz, and rock fragments (phyllite, limestone, quartzite) and a natural clay binder. The production process of the clay, based on the composition and the petrographic analyses, was established to entail firing at low temperatures, below 600 °C [14].

The sample from Tepe Narenj showed a relatively soft-fired clay, which tends to disintegrate with the handling. The samples was made up of a red clay with bright sand (feldspar, quartz) and rock fragments (phyllite, limestone, quartzite). A natural clay and fine-grained calcite served as binders. The peculiarity of the sample is that relatively large crystals of gypsum are still present. As gypsum is thermally not stable during a normal, high temperature firing process, it can be concluded that this mineral is either the result of a secondary process, or that the firing temperature was very low. Chemical analyses of phyllosilicates show that most of them were altered during the firing process. The compositions is quite rich in calcium, in contrast with the naturally occurring ones [14]. The rock and mineral fragments could derive from sedimentary rocks (limestone) or from crystalline rocks (hornblende, K-feldspar, garnet). The composition of the mineral hornblende found in this sample is different from that found in the sample from Tapa Sardar, indicating a different region of provenance for the sand.

4.1.3. Stucco

Recent technical studies of stucco supports for the architectural decorations of Buddhist buildings and artworks (sculptures) showed different stucco mixtures [14,36], always comprising a calcite matrix [17,36]. In a few cases, the analyses on stucco samples of architectural decorations and on museum artworks showed the presence of gypsum [17,36–38]. Only in the cases of architectural decorations were fibre-like pore structures found. Vegetable fibres may have been added to improve the stucco application on irregular masonry, such as those found in Gandharan Buddhist sites, and, possibly, to slow down the drying, preventing the formation of cracks. This technique was typically used on painted walls in the Ancient Middle East [39,40].

4.2. Ground/Preparation Layers

Ground layers for polychrome decoration were used on all types of supports. Sometime only the ground layer is still preserved today on the surface of the artworks, without the coloured decoration. At times, polychromy as well as the ground layer are no longer visible, because of a higher state of degradation, or because the remaining traces been accidentally removed during cleaning.

The composition of the ground layers is different, depending on the support:

- schist artworks showed a white ground layer containing a high amount of calcium carbonate and aluminum-silicates [17];
- a thin ground layer is often visible on stucco artefacts and architectural decorations, at times covered by a thin polychrome layer applied to improve the surface smoothness [14,17,36–38,41]
- clay artworks always present red coloured ground layers of variable chemical composition [17].

4.3. Pigments

The presence of traces of polychromy was well known in the 20th century [37], although scientific investigations were scarce [8], and mostly focused on Afghan sites [42–44] and a few stucco artworks [41].

Our team systematically carried out scientific investigations on polychromy on all types of supports. Data show that the most frequently used colour on polychrome sculptures and reliefs on different supports (stone, stucco and clay) and on stucco coatings of Buddhist monuments [8] is red [14,17,19,36]. The prevalence of the use of red, in various shades, was also highlighted on Parthian Iran stucco decorations [45,46], and Parhtian Nisa sculptures [47], on mural paintings from ancient sites of Central Asia [48,49], in Indian sculptures and reliefs [12,50,51] and also in Greek and Roman art (see e.g., [52–54]). In the majority of the cases, the red colour is based on a red ochre, which is characterised by great stability over time [14,17,19,36]. A red-orange pigment was also employed on Gandharan stone, stucco and clay sculptures based on lead, most likely minium, a red oxide, in some cases used a coloured layer under the gold leaf on stone artworks [17]. In a few cases, vermillion, mercury sulphide, was found on clay statues [17]. Ochre pigments were frequently used also in yellow hues, both on stucco and clay sculptures [17] and orpiment, an arsenic sulphide, was found on clay statues [17]. Black pigments are present on clay statues and were obtained from calcined bone. Lead white was found on the ground layers of stone sculptures and reliefs and pigment on clay statues [16,17]. The same red, yellow, white and black pigments were also used in ancient Greek and Roman art (see for example [54–57]) and in Indian [51,58–60], Iranian [45,46] and Central Asian [47,49,61] wall paintings and artworks, as well as on Chinese sculptures [62].

In the Gandharan artworks analysed, the blue colour was always based on lapis lazuli, which can be mined locally [63]. The same pigment was also found in wall paintings in Central Asia [49]. No copper pigments, such as azurite, used on Tutang Buddha Sculptures in Taiyuan (China) [62], nor Egyptian blue, widespread in the ancient Mediterranean area (see e.g., [64–68]) and in Parthian Iranian and Nisa stuccos [45–47,61], were found. Green was rarely preserved on Gandharan artworks, but probably used, as it has been found on wall paintings and on stucco decorations of Iranian buildings—earth green pigment—[45,46], and in ancient settlements of the Central Asia -atacamite—[49] and on Indian wall paintings (e.g., [58,60]) and Chinese artworks—atacamite—[62]. Probably its absence, or limited presence, also noted on Indo-Afghan stuccos [37], may be ascribed to degradation phenomena. According to some scholars though, the scarce use of green was perhaps due to its absence in the palette of the five main colours mentioned in some Sanskrit texts [48], written in more recent times than the Gandharan period, but referring to a more ancient artistic tradition [50,51].

In some cases, differently pigmented layers were applied on top of each other, most likely to obtain specific shades of colours [17], as it was customary on Roman polychrome sculptures (see for example [66,69,70]) and on Indian [37] and Chinese artworks [62].

A total of 19 samples have been chosen for binder analysis, representing the different types of objects investigated: 6 samples were collected from polychrome statues coming from museum collections, and 13 samples were collected from architectural decorations, 70% of which were sampled directly at the archaeological sites in Pakistan, while the remaining 30% derived from objects belonging to museum collections. GC-MS and proteomics have been used in a complementary way: 6 samples were studied by proteomics while 14 were analysed by GC-MS. A summary of the analyses performed, samples analysed and the data obtained from each sample are presented in Supplementary Materials Section S4. Table 3 summarises the results.

Table 3. Summary of Gandharan art binder analysis results (n.d. non detected, i.e., below the LOD).

Sample	Ref	Century CE	Provenance	Statue	Architectural Decoration	Support	GC-MS	Proteomics
ex-MNAO 1 (Rome Museum) inv.1240	[16]	ca. 2nd–3rd	Butkara (Pakistan)		X	stucco	traces of proteinaceous material (present, below LOQ)	
ex-MNAO 1° (Rome Museum) inv.1240	[16]	ca. 2nd–3rd	Butkara (Pakistan)		X	stucco	traces of proteinaceous material (present, below LOQ)	
ex- MNAO 5A (Rome Museum) inv.4423	[16]	ca. 1st–2nd	Panr (Pakistan)		X	stone	n.d.	
ex-MNAO 7A (Rome Museum) inv.2519	[16]	ca. 2nd–3rd	Butkara (Pakistan)		X	stone	n.d.	
ex-MNAO 11A (Rome Museum) inv. 435	[16]	ca. 3rd–4th	unknown	X		stucco	traces of proteinaceous material, (present, below LOQ)	
AKD14C	[18]	late 3rd	Amlukdara (Pakistan)		X	stucco	traces of proteinaceous material, (present, below LOQ)	animal glue egg
BKG1123A	[18]	3rd	Barikot (Pakistan)		X	stucco	traces of proteinaceous material, (present, below LOQ)	
BKG1123B	[18]	3rd	Barikot (Pakistan)		X	stucco	n.d.	
MG18957 (Museum Guimet of Paris)	[18]	7th	Fundukistan (Afghanistan)	X		clay	-	animal glue milk egg

Table 3. *Cont.*

Sample	Ref	Century CE	Provenance	Statue	Architectural Decoration	Support	GC-MS	Proteomics
MG18959 (Museum Guimet of Paris)	[18]	7th	Fundukistan (Afghanistan)	X		clay	-	animal glue milk egg
MCM4 (Civic Archaeological Museum of Milan)	[18]	2nd–3rd	unknown	x		stone	-	animal glue
MCM5 (Civic Archaeological Museum of Milan)	[18]	4th–5th	unknown	x		clay	-	animal glue
MCM25 (Civic Archaeological Museum of Milan)	[18]	unknown	unknown	x		clay	-	animal glue
GBK (18)	present work	2 nd–3rd	Gumbat/Balo kale (Pakistan)		X	stucco	animal fat	-
GBK (17)	present work	2nd–3rd	Gumbat/Balo kale (Pakistan)		X	stucco	animal fat	-
BKG 1123 (15)	present work	2nd	Barikot (Pakistan)		X	stucco	traces of proteinaceous material, (present, below LOQ)	-
A3	present work	2nd	Gumbat/Balo kale (Pakistan)		X	stucco	n.d.	-
C11	present work	3rd–4th	Amlukdara (Pakistan)		X	stucco	egg, animal glue	-
B8	present work	3rd	Barikot (Pakistan)		X	stucco	egg, animal glue	-

All analyses carried out so far related to the detection of proteinaceous binders, indicating the use of a tempera technique, in agreement with the historical sources and the few analyses performed on Central Asian artefacts [71–74]. Silpa defined the Asian painting technique as a tempera, that is, a painting technique in which the pigment is dispersed in a water-based medium, but scientific data confirming this affirmation is still necessary in most cases [75].

In Gandharan art, analyses always show the presence of trace amounts of proteins, at times not sufficient for a quantitative analysis, which is necessary for a reliable identification of the source of the material. This is expected in analyses of traces of polychromy found on archaeological objects and buildings, as the original paint layer has undergone centuries of ageing. In the region of interest, environmental conditions definitely entail exposure to quite extreme temperature conditions, often including repeated cycles of freezing, fusion and evaporation of moisture, which affect the mechanical stability of the paint layers, and thus, the preservation of the binder [76]. On the other hand, the detection of small amounts of protein markers allows their derivation from recent conservation treatments to be confidently excluded, as a conservation layer made of proteins would definitely lead to completely different amounts of proteins in the samples analysed.

Both proteomics and GC-MS analysis performed on samples from Gandharan art highlight the use of animal glue in most of the samples and of egg in some cases [17,18]. Animal glue seems to be mainly related to statues and clay figurines preserved in museum objects, while egg mainly occurs in architectural decorations on stucco. Milk was detected only in few cases.

The identification of animal glue is quite straightforward: the detection of hydroxyproline by GC-MS or of peptides with the repeating XaaYaaGly sequence (with Xaa and Yaa being mostly Pro and Hyp [77]) by proteomics make the detection of a collagen-based material unambiguous. Proteomics allowed the identification of animal glue in all the samples analysed. Several peptides corresponding to collagen from Mammalia were identified in all cases. Moreover, the origin of collagen was identified as derived from Bovidae species (sample MG MG18957). Animal glue has been widely used for centuries in Europe as a binder of preparation layers, and its use in Asian ancient polychromy is also reported in written sources [73] and documented, both on polychrome objects and mural paintings from China [73,78–86], the Himalayan [87] and Afghanistan [88]. Animal glue was also used in polychromies of the Mediterranean area since very ancient times [89].

Animal glue presents a certain sensitivity to humidity. This might explain why the objects showing the presence of animal glue are sculptures which were probably intended for indoors. It is necessary to highlight that the samples analysed by GC-MS often showed the presence of hydroxyproline, but a chromatographic profile which is not typical of animal glue, indicating that collagen is a minor component. Proteomics analyses show the reliable identification of a small number of peptides from collagen. There are a few hypotheses which could explain such analytical behaviour. Animal glue could have been used as a thickener of the main paint binder, and thus added in very small amounts. Another possibility is that a diluted solution of animal glue could have been used as a sizing agent, which, given the high porosity of most of the substrates investigated, would not have left a visible layer, but may have penetrated the substrate. Also, animal glue could have been used as an organic additive of stuccoes, and thus, again, would be present in small amounts [90].

Egg yolk was identified in a few samples by GC-MS, and by proteomics in only one sample. The identification of egg in aged and degraded samples is quite challenging, given the high level of glycosylation of egg proteins, which hampers an efficient digestion into peptides [91]. Egg yolk or whole egg produce both a very resistant paint layer, which is well suited to externally exposed polychromy. Egg though produces paint layers which are quite thin, and we cannot exclude that other water-based binders, such as polysaccharide gums or animal glue were added to modify the rheological properties of the paint.

Unfortunately, despite tentative separation of the paint and preparation layers, we do not have enough data at present to reliably assess which materials are associated to the paint layers and which to the preparation layer. An interesting sample analysed in this context though, is a sample collected from a solidified drop of paint (Supplementary Materials Section S4). The principal component analysis of the amino acidic profile points to the use of egg as binder (Supplementary Materials Section S4) confirming the use of this binder in the coloured paint layers.

The use of egg as a paint binder is documented from very ancient times, detected in the paintings from the Mediterranean Area since the Bronze age [88,92–94], and in polychrome objects spanning from the Mediterranean Sea [95] to China [78,81,82,85,96], though the Silk Road [88].

Milk was found in only one of the samples analysed. Its use as paint binder in the Ancient World is less documented [78], although it was identified in the Buddhas of Bamiyan (Afghanistan, 7th cent. CE), probably used in both original and historical restoration layers [88].

A polysaccharide gum—tragacanth gum—was identified in Gandharan art only in one sample consisting of a clay support showing a blackish layer on top [17,18], whose binder was found to be proteinaceous, showing similarities with the Buddhas of Bamiyan, where polysaccharides were used as insulation layers over the clay support [88].

It is at this point necessary to make a very important consideration. The identification of a paint binder is based on the comparison of the experimental data—peptide MS/MS spectra in the case of proteomics and quantitative amino acid profiles in the case of GC-MS—to a database of reference samples. The choice of the database is thus absolutely fundamental to allow a reliable identification of a material. Although the paint binders in the samples are definitely based on proteins, we cannot exclude that in addition to those detected, or instead of some of those detected, other protein-based materials may have been used. More research is needed on Asian polychrome artefacts to better understand the possible classes of materials used, and thus improve our understanding of the technologies and painting techniques.

5. Conclusions

The research described herein aims to shed light on the materials and techniques used by the Gandharan painters of the 2nd to 7th century CE, and to understand the influences exerted on them by Eastern and Western artistic traditions.

The use of a tempera technique based on proteinaceous-based aqueous media has been evidenced for all types of supports and objects (architectural decorations, reliefs or statues) with egg and animal glue being the main binders identified.

The supports of Gandharan artworks (statues, reliefs and architectural decorations) are stone, stucco and clay, presenting in the majority of the cases a ground layer of varying composition depending on the substrate. The colour palette is wide, mainly based on red and yellow (mostly ochres), white (lead white), black (bone black), and blue (lapis lazuli). The pigments analysed, with the exception of lapis lazuli, which was mined locally, are common to those used also in the Mediterranean area, in Central and East Asia. The red colour is the most common on all artefacts.

Data show that the Gandharan artistic techniques present common features to the polychromy of the Ancient World, spanning from the Mediterranean region to China, along the Silk Road. More research is still necessary to determine the influence of Hellenistic or Roman cultures, due to the continuation of Alexander's cultural legacy through Indo-Greek reigns and trade with the Mediterranean region. This theme is fundamental to better understand the cultural dynamics in which Gandharan art developed, highlighting artistic and technological influences of and/or parallels with populations that had cultural and commercial exchanges with this Himalayan population.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/heritage5010028/s1. Section S1. Arhchaeological description of the Samples [97]; Figure S1. Sample BKG 1123, 16B: polychrome stucco architectural decoration of building with traces of red colour from Barikot (© ICR, photo Edoardo Loliva); Figure S2. Sample GBK (18), stucco architectural decoration of building from Gumbat/Balo Kale, photo of cross section (Dinolite, 20×): the white ground layer is visible; Figure S3. Sample GBK (17), stucco architectural decoration of building from Gumbat/Balo Kale, photo of cross section (Dinolite, 20×): the white ground layer is visible. Section S2. GC/MS quantitative analyses; Section S3. GC/MS limits of Detection and Quantitation; Section S4.: Binder analysis of samples from Gandharan polychromies; Figure S4. Statistical data related to the samples analysed and the type of analysis performed; Table S1. Relative amino acid percentage content of the samples with an amino acid content above the quantification limit; Figure S5. PCA score plot of the sample consisting in a drop of paint.

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