Schists and Pigments from Ancient Swat (Khyber Pukhtunkhwa, Pakistan)

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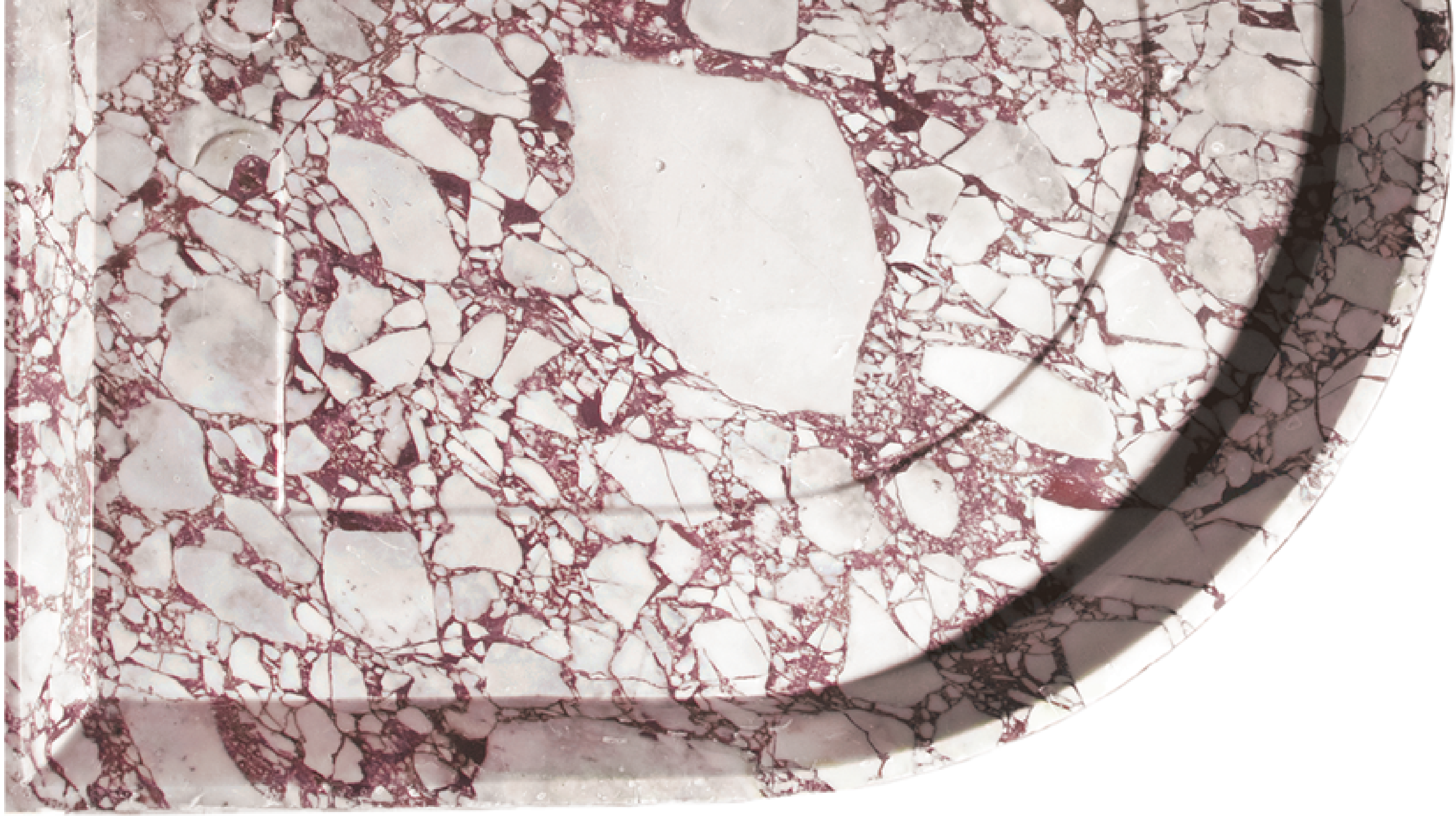

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CONTENT

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SCHISTS AND PIGMENTS FROM ANCIENT SWAT (KHYBER PUKHTUNKHWA, PAKISTAN)

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Abstract

Since the earliest agricultural communities settled in the greater Indus basin in the 7th millennium BC, rocks and minerals have had an important role in the local processes of social evolution in order to signal the superior status of elites, as Italian excavations and surveys demonstrated in numerous projects (IsMEO-IsIAO and National Museum of Oriental Art in Rome, MNAO).
This paper presents results from minero-petrographic analysis carried out from sculptures exposed in MNAO, with the aim of better understanding their context and provenance.
The petro-textural characters have implications for both the conservation of the sculptures and reconstruction of the provenance history of the raw materials from the Swat Valley.
A set of brightly coloured particles made of different raw materials (pigments and gold leaf) was also analysed. These precious materials were probably placed in the reliquary alongside a few cremated bones in the frame of a highly symbolic, ritual setting.

Keywords
schist, ancient Swat, reliquary, pigment, gold leaf

Introduction

In the framework of an ongoing study on the collection of Gandharan stone sculptures at the National Museum of Oriental Art (MNAO), Rome, renewed attention is being paid to the characterisation of the raw materials and the state of conservation of some artworks. The new investigations follow several excavations (Fig. 1) performed after insights arising from pioneer studies and geological surveys[1] at the Swat District of the Khyber Pukhtunkhwa province (northern Pakistan).

1 FACCENNA 1974, 126-176; DI FLORIO *et al.* 1993a.

The Gandhara sculptures from the MNAO are carved from metamorphic rocks belonging to the ophiolite sequence of the Indus Suture Zone. Although these rocks are commonly given general term of "schists", they include a large variety of metamorphic lithotypes with different mineral assemblages and different petrographic fabrics. The identification of the petrographic properties of these sculptures is crucial for understanding the lithology and for providing insights into the provenance of the raw material. Whereas the scientific characterisation of ancient marbles is one century old[2], the first mineralogical investigations on the "metamorphic schists" used in Gandharan sculptures begun less than 50 years ago[3]. Therefore, identifications are still scarcely supported by a specialised geological know-how. New information on the lithotypes on record is considered in the light of the local outcrops and the general archaeological and cultural contexts of the sculptural complexes.

Geological setting

The geological history of Pakistan[4] is connected with the continental collision between the Indo-Pakistani and the Eurasian plates, starting from the Cenozoic[5]. The continental collision after the consumption of the former interposed Tethyan oceanic floor[6] resulted in a prolonged phase of tectonic stacking of oceanic-derived units. The southern margin of the Neo-Tethyan seaway, which rimmed the Indo-Pakistani subcontinent, was incorporated into the Indus Suture Zone, together with major island arcs (such as the Kohistan island arc terrane)

2 MARIOTTINI 1998, 23-35.

3 CURTOIS 1962, 107-113.

4 GAETANI *et al.* 2004, 56.

5 MOLNAR *et al.* 1977, 30-41.

6 KHAN *et al.* 2009, 366-384.

Fig. 1. Snapshots of excavations (elab. by B. Mazzone)

and volcano-sedimentary orogenic wedges[7]. The Khyber Pakhtunkhwa Province of Pakistan is dominated by the Kohistan-Ladakh tectonic complex (Fig. 2), a segment of the Indus Suture Zone developed during the northward underthrusting of the thick Indo-Pakistani shield lithosphere, with southward piling of the tectono-metamorphic units[8]. The metamorphic units include a heterogeneous sequence of intermingled HP/LT (mainly in the blueschist facies) and MP/LT (in the greenschist facies) rocks characterised by specific mineralogical assemblages[9]. All these lithotypes are presently exposed along the Indus Suture Zone. In the Swat District, decametric- to hectometre-thick lenses of talcschists and serpentinites are interdigitated with epidote-rich greenschists, magnetite and muscovite-bearing chloritoschists, marbles and phyllites. The top of the sequence generally includes a metapelitic unit (the Saidu Unit) composed of garnet-kianite-staurolite micaschists, paragneiss, amphibolites, prasinites and chloritoid-bearing calcschists[10].

Presently, the structural setting of the region, including the structural boundaries between the tectono-metamorphic units, is defined by the brittle deformation network, which consists of the imbricate thrust splays along which the considerable crust shortening was produced.

Material and quarries

In the Swat District, several quarries are localised in the immediate proximities of sacred areas[11]. These quarries correspond to the exposures of the ophiolitic rocks from the Indus Suture Zone. The large variety of lithotypes from the ophiolite sequence represented a great wealth of available materials adopted for both carving and building construction. These lithotypes mainly correspond to chloritoschists, talchists, and serpentinites. These rocks are characterised by plano-laminated foliation that provides preferential cleavage planes to the rock that can be well worked during carving. Chloritoschists and talchists were used for many sacred sculptures and petroglyphs coming from the Gandharian sacred areas[12]. The talcschist lithotype,with its finely schistose fabric, was used at least in a lion sculpture in the Butkara site. The wall blocks of this site were built with the same talcschist. It comes from many mines situated in several valleys of Khyber Pakhtoonkwa, like Jamrud, Hazara, Shangla, Parachinar, etc., even also

7 DI PIETRO *et al.* 2008, 1428-1440.

8 PETTERSON 2010, 287-327.

9 KAZMI, JAN 1997, 554.

10 TAHIRKHELI 1982, 1-51.

11 DI FLORIO *et al.* 1993a; DI FLORIO *et al.* 1995.

12 OLIVIERI, VIDALE 2004, 121-180.

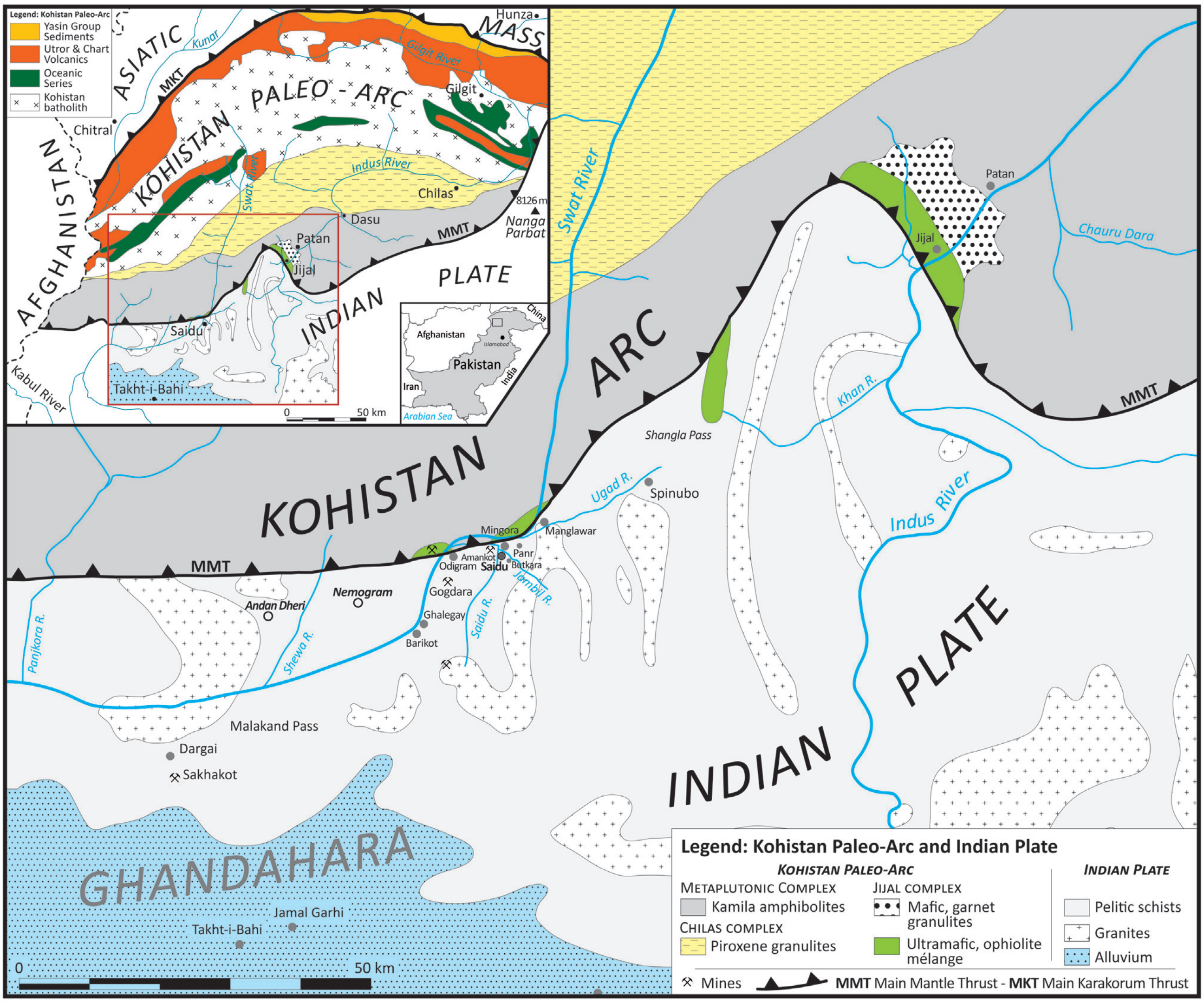


Fig. 2. Tectonic map of the Swat Valley (after FACCENNA *et al.* 1993, 257-270). The geodynamic scenario shown in the inset is after TAHIRKHELI (1982, 1-51) and PETTERSON (2010, 287-327)

used for the stability of the pigments or for simple cosmetics as in the centres of Hindu[13].

In addition to sculpturing, the ophiolitic rocks were also used as quarrying material for building. The ophiolitic rocks of Mingora were identified as the extraction area on the right side of the Swat Valley, but also on the left side of the same valley, opposite the town of Saidu Sharif. However, just up the more recent hills, intense modern mining for emeralds has cancelled every trace of excavations[14].

In sacred areas, particular containers were found during archaeological surveys. These containers, having a shape similar to small cups, were probably devoted to multiple uses, like to prepare plasters or "stucco", to crush materials, or as cavities for food preservation. In many cases, these containers were used for baking lime in sacred areas, as observed in Butkara, Saidu Sharif, Panr and more recently in Amluk-dara[15].

In ancient time the reliquaries contained relics of Buddha with a shape like a 'stupa' as in the Swat Region at Kanishka (Peshawar), Barikot, which recalls the function of the objects. Hindu and many other religions exhibit reliquaries in shrines or temples. Often organs or bone fragments of 'saints' can be separated from the main burial site and recovered in holders engraved with precious metals and rare stones. Not only do the colours have a high symbolic significance, but a stupa is an important form of a reliquary in Buddhism. Finally, there are also many sources of gold over all the area[16].

13 VIDALE, KENOYER 1992, 92-95.

14 SNEE *et al.* 1989, 93-123.

15 OLIVIERI 2006, 137-155.

16 KHAN 2004, 27-40.

Samples	*Petrographic classification*
MAO4423, MAO2464, MAO2158, MAO4059, MAO2244, MAO1117, MAO4172, MAO2158, MAO4153	Serpentinite
MAO1304	Serpentinite with calcite
MAO4244, MAO1169, MAO4152, MAO2519, MAO1134	Schistose serpentinite
MAO 2080	Talcschist with antigorite

Table 1. List of selected samples and petrographic classification

Petrography of selected samples

The Gandhara finds were preliminarily analysed[17] under the supervisor of the ISMEO. Within this section, we report on a list of the main petrographic characteristics of selected artefact samples stored at MNAO, by focussing on (i) the description of the main micro-structural rock properties, (ii) the definition of principal and secondary mineralogical aggregates, and (iii) the petrographic classification of the samples.

The petrographic analyses were performed on 16 polished thin sections (30 µm thick) taken from the sculpture samples (Table 1). Observations were made using an optical transmitted light microscope equipped with 2x, 4x, 10x, 20x, and 40x magnifications and digital camera acquisition.

At the sample scale, most of the samples have a homogeneous green-grey colour over the entire surface of the investigated thin section. By contrast, sample MNAO2080 shows a homogeneous whitish colour. From a textural point of view[18], these samples show either a sub-millimetric-thick foliated fabric or a massive fabric.

The petrographic characteristics unravelled at micro-scale allow classifying the serpentinite as the predominant lithotype, with schistose serpentinite and talcschist as subordinate lithotypes.

Serpentinite samples are characterised by a dominant massive fabric defined by the random orientation of the main mineralogical assemblage (Fig. 3a). The massive fabric is often crosscut by a system of microveins not showing a systematic trend. Antigorite is the main mineralogical constituent, providing up to 60% of the main fabric investigated at the thin section scale. Antigorite grows both as lamellar and fibrous-acicular crystals. Fibrous antigorite occurs dominantly in the investigated samples (e.g., samples MNAO2464, MNAO 2158). Fibres appear either isolated or in aggregates to form fan structures. Secondary mineralogical species are mainly represented by white mica (crystals having up to 1 mm in length), calcite (fine-grained crystals in micro-veins together with antigorite), and chlorite (very fine-grained crystals within the groundmass). In detail, white mica appears as single crystal that is (i) either incorporated by the antigorite groundmass, or (ii) superimposed on the antigorite groundmass. In the first case, white mica crystal shows irregular habitus, folded cleavage, and faint chemical alteration along the crystal rims; however, in the second case, white mica presents well-defined, unaltered, habitus. Accessory minerals are magnetite and epidote (up to 20 µm in size), chloritoid (up to 70 µm in length), and rutile. Chloritoid occurs as single tabular crystal with corroded shape (sample MNAO 1117). Rutile appears as mineralogical relict mantled by the antigorite groundmass.

Sample MNAO 1304 corresponds to a serpentinite with massive fabric and diffuse crystallisation of calcite aggregates. Antigorite (and subordinate, very fine grained chlorite) is the main mineralogical constituent. Calcite crystal has a size over 100 µm and does not show relation of textural equilibrium with antigorite crystals, suggesting that calcite is a late mineralogical phase developed after the antigorite groundmass.

Schistose serpentinite samples show an internal fabric consisting of penetrative, few µm-thick foliation (Fig. 3b). The foliation is made of aligned fibrous antigorite, up to 300 µm in length (Fig. 3c). Within the foliation, secondary and accessory minerals are dispersed. These correspond to rutile, magnetite, chlorite, and white mica up to half a millimetre in size. Rutile and magnetite often show corroded rims, while white mica may appear with regular habitus.

Talcschist is characterised by the development of an incipient foliation defined by faint alignment of talc crystals (Fig. 3d). Talc crystals are acicular to lamellar in shape, up to 100 µm in length. Foliation is up to 30 µm in width, with local deformation in microfolds. Within the foliation, acicular to fibrous antigorite occurs as a relict mineralogical phase transposed by the main foliation containing talc.

Also the inquiries with the investigations at the scanning electron microprobe (SEM-EDS) scale confirm the petrographic observations, evidencing the occurrence of mineral phases rich in Al_2O_3 (chloritoid) besides quartz and oxides of Ti (especially rods of rutile and aggregates of ilmenite), Fe (magnetite), often associated with minimal quantities of Mn, Cr and V. Most of the analysed

17 FACCENNA *et al*. 1993, 257-270.

18 SPRY 1969, 350.

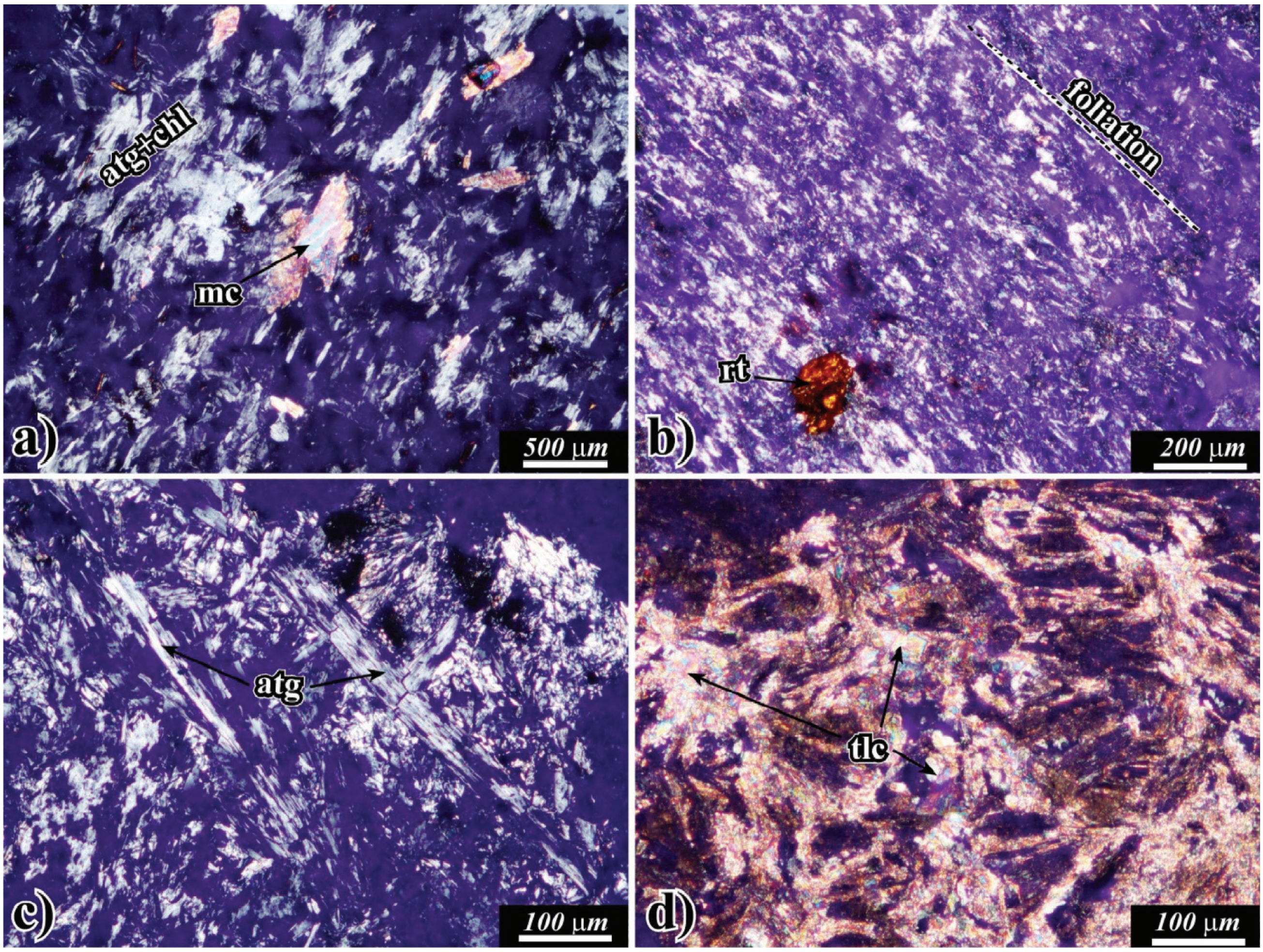


Fig. 3. Petrographic features of selected samples at thin section scale. (a) serpentinite with massive fabric defined by antigorite (atg) and fine grains of chlorite (chl) in the groundmass, with late crystallised grains of white mica (mc); (b) serpentinite schist characterised by penetrative foliation marked by aligned fibrous antigorite incorporating relict mineralogical phases like rutile (rt); (c) detail of the serpentinite schist fabric evidencing fibrous antigorite; (d) talc aggregates (tlc) in microfolded fabric of talcschist. All pictures have been taken at crossed polars

samples show the occurrence of Zr and REE such as lanthanoids (Sm, Gd, Dy, Er, ecc.). The image and its spectrum of the section of the sample MNAO 2158 (Fig. 4) show also the presence of a granule cerium, neodymium and lanthanum, to be referred to monazite-(Ce): (Ce, La, Nd, Th)PO_4. Further, it is possible to find small grains with variable amounts of zirconium (zircon), titanium (rutile), or iron (flaky or platy of ilmenite) forming single acicular crystals or frequently giving forms at a sharp angle, called knee-shaped twins. In some findings, the zircon and the rutile have plaques of ilmenite and magnetic alteration with hematite (Fig. 5, sample MNAO 1148.). The silico-aluminate phases also have problems of chemical and textural alteration (i.e. decussate texture) regarding the antigorite, the chloritoid, epidote and muscovite.

Finally, according to the examinations done at the microprobe scale, SEM-EDS analyses give information about the chemical characteristics of the groundmass. In particular, it has been possible to verify that approximately the same concentration percentage in weight of the elements (Mg, Al, Si, Fe) occurs between grains and the surrounding groundmass.

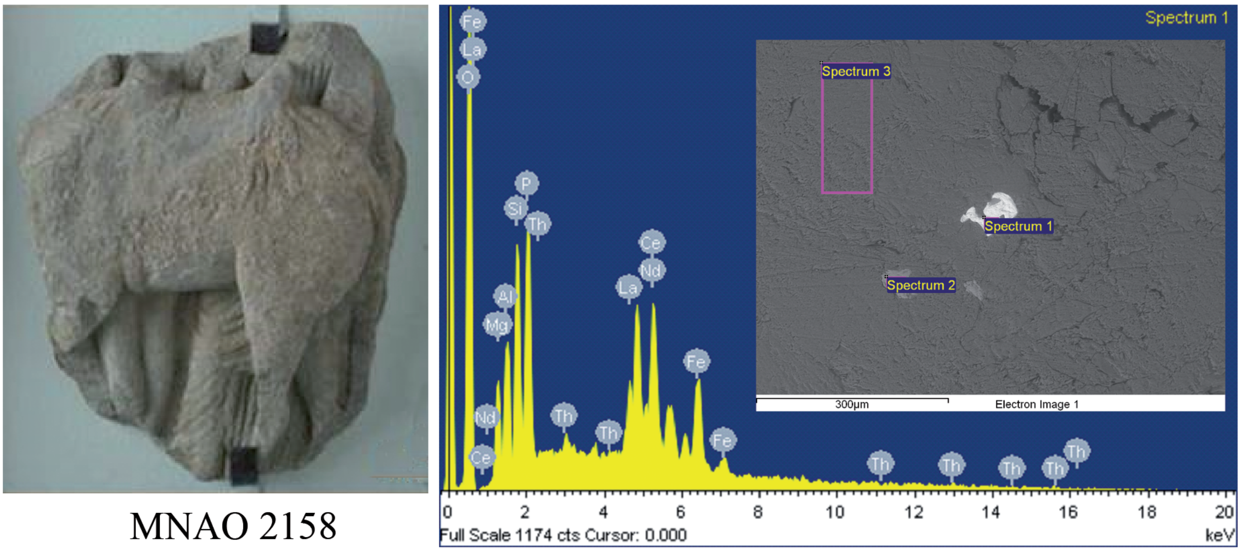
Painting and pigments

Analysis of schists[19] provides reliable and significant data on the nature of the colours, the preparation layer and about the manufacturing process.

Following the previous stereoscopic microscope and UV/vis observations[20], XRD analysis integrated with SEM-EDS show that natural pigments (e.g. red and white) were readily available in the areas surrounding the stone workshop.

19 Based on an unpublished report from MARIOTTINI 2014, 7.

20 STERLING GLEASON 1960, 244.



Processing option : All elements analysed (Normalised). All results in weight%

Spectrum	In stats.	O	Mg	Al	Si	P	S	Ti	Fe	La	Ce	Nd	Th	Total
Spectrum 1	Yes	30.49	3.79	4.03	5.90	7.48			7.15	8.54	20.50	10.43	1.69	100.00
Spectrum 2	Yes	49.76	2.58	2.47	3.41		0.13	35.48	6.16					100.00
Spectrum 3	Yes	48.76	8.29	8.99	13.25		0.26		20.46					100.00
Max.		49.76	8.29	8.99	13.25	7.48	0.26	35.48	20.46	8.54	20.50	10.43	1.69	
Min.		30.49	2.58	2.47	3.41	7.48	0.13	35.48	6.16	8.54	20.50	10.43	1.69	

Fig. 4. Spectrum and occurrence of Monazite-(Ce), (SEM sample M2158)

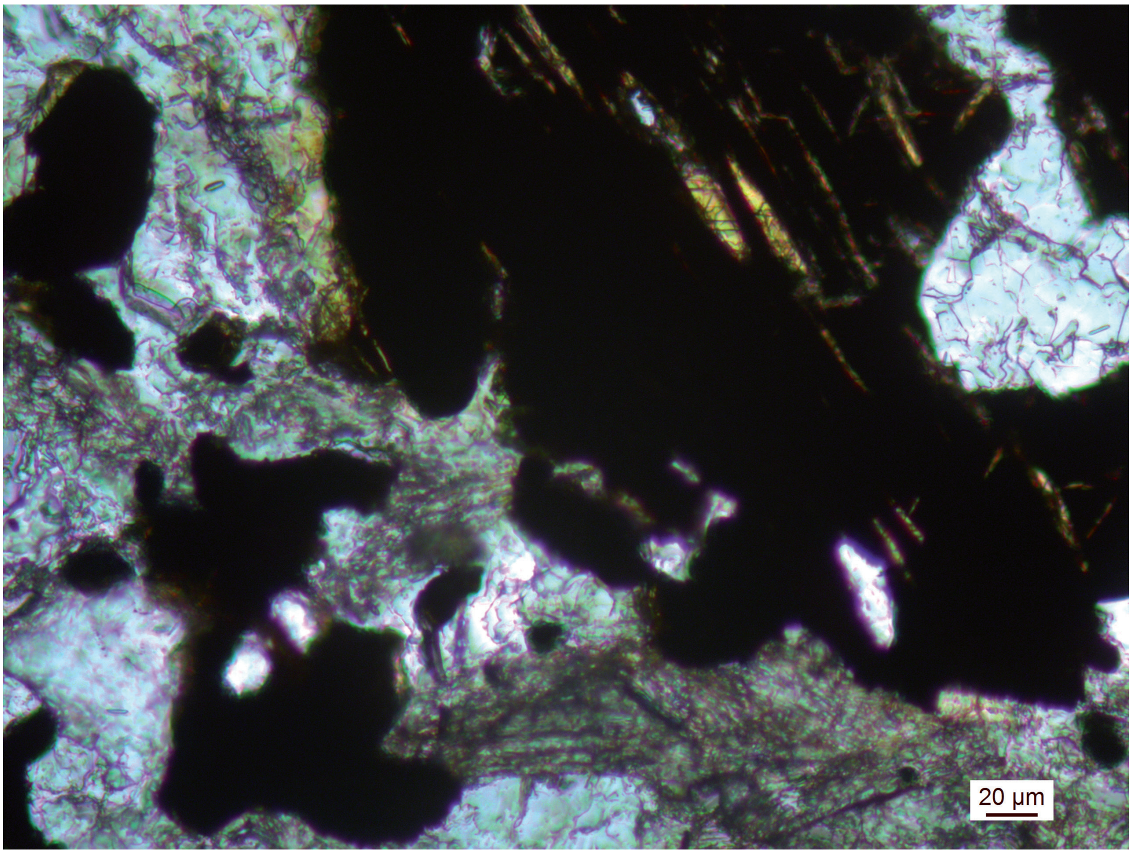


Fig. 5. M1148 - Magnetite partially rimmed by rutile, altered ilmenite-leucoxene, titanite and small zircons (crossed polars)

Nevertheless, due to their textural properties and the effects of dangerous washing during excavations, the traces of the colours on green-schist have less resistance to weathering than those on marble (i.e. 'fresco' techniques).

Many red grains sampled from the schists are assigned to ochres, whereas white grains refer to the so-called calcium carbonate or calcite (see MAO1169bind_whi, Fig. 6).

Sometimes the coloured grains are accompanied by silicates (e.g. talc) or serpentinised rocks (chlorites or amphiboles), which are characterised by a complex internal structure[21].

A singular composition has been observed in an artefact coming from Panr, where 'biacca' was used (Fig. 6, sample MAO4423).

Preliminary investigations on 'stuccos' (Fig. 7) show that their colours are probably similar to the ones on stones, such as the red ochre, with comparable quantities of chemical elements like Mg, Al, Si, and Fe-Ti.

Interestingly, sample M1293 is characterised by small particles of gold leaf folded in various shapes (Fig. 8). The same sample exhibits a fine crystallization of azurite on compact and banded malachite (Fig. 9). The alloy is characterised by Au-Ag, whereas SEM analysis of small opaque areas show high values in Fe-Ti, Al, Si, alkali and alkaline elements.

Workability, alteration and conservation problems

The green rocks of the Swat have workability and conservation problems partially known for various kinds of lithotypes. The conservation problems cannot be reviewed only by studying the final shapes of their products. In fact, the alteration comes from far away and often depends on their previous story, until the processing and the conservation in the final destination's environment[22]. However, in general the mineralogical and textural component is not a particularly difficult factor for the sculpture, even for lithotypes with an abundance of magnetite, sometimes altered and with limonitic residues. Actually, the soapstone itself, for its softness can be as easily carved as wood. For the micaschists, with a bigger grain than the phyllites, the freedom of handling becomes more crucial with the loss of material in the spalling; the phenomena of detachment (i.e. sample 4152) should be stabilised to avoid the loss of readability of the artifacts in the future.

Regarding the conservation problems, of a more specific mineralogical kind, the rocks of the Swat, notoriously refractory like other green rocks, have proved to be usually resistant enough to chemical alteration. The practical note of soaking the findings in acid solutions (widely practiced in non-scientific contexts) in order to remove veils and carbonate concretions, has still some very crucial aspects. In fact, when the original rock (even if rich in minerals resistant to the acid attacks, like talc) contains mineralogical phases made of different percentages of apatite, brucite, chrysotile, monazite and obviously calcite and dolomite, it will have a low mechanical resistance to weathering, because it has proved to be sensitive to carbonic acid.

Conclusions

According to the inquiries made during this research on some of the Gandhara artifacts of the MNAO, only some lithotypes have more or less traces of mineral chloritoid, which is very common in the materials of the Gandhara arts according to some studies[23]. Moreover, no quarries of the chloritoid-bearing phyllites often used for the Gandhara statues have not been documented. In fact, the majority of the statues have been sculpted in phyllites; others in green schists and only a few in talc schist (see *soapstone*). The phyllites of the statues studied[24], and other researches[25] have similar mineralogical and rheological aspects, but the chemical composition of the chloritoid widely varies[26]. According to surveys, we believe that one of the best areas to control the phyllites used for the statues of the Swat is the middle valley of the Jologram, especially on the south bank where fine and gradated phyllites of chloritoid come to the surface.

Petro-mineralogical insights from this study help to classify phyllites, at least some of these, as serpentinites, schistose serpentinites, and talcschists. Further basic inquiries have to be completed in the next step

21 HOWIE 1982, 1155-1178.

22 MARIOTTINI, VIGLIANO 2013, 219-233.

23 KEMPE 1982, 25-28. The author confirms the presence of schists rich in Al with a paragenesis in chloritoid-paragonite-muscovite-quartz, compatible with the formations mentioned above, but not yet discovered on the field as sure proofs of ancient extractions. The question of the chloritoid is crucial, because the green schists with such amount of phyllosilicate are common in metamorphic environments of orogenic overthrust but often in regions out of the Swat, far beyond the Khyber Pass, in the middle of the Afghan territory. According to other scholars (CAMBON, LECLARIE 1999) the materials used in Gandhara art, are very homogeneus and "the only exceptions are related to Swat, with chloritite as a case apart..".

24 See also KEMPE 1986, 79-87.

25 NEWMAN 1984, 288; NEWMAN 1992, 163-174; REEDY 1992, 241-287; REEDY 1997, 267-283.

26 NEWMAN 1988, 120-131.

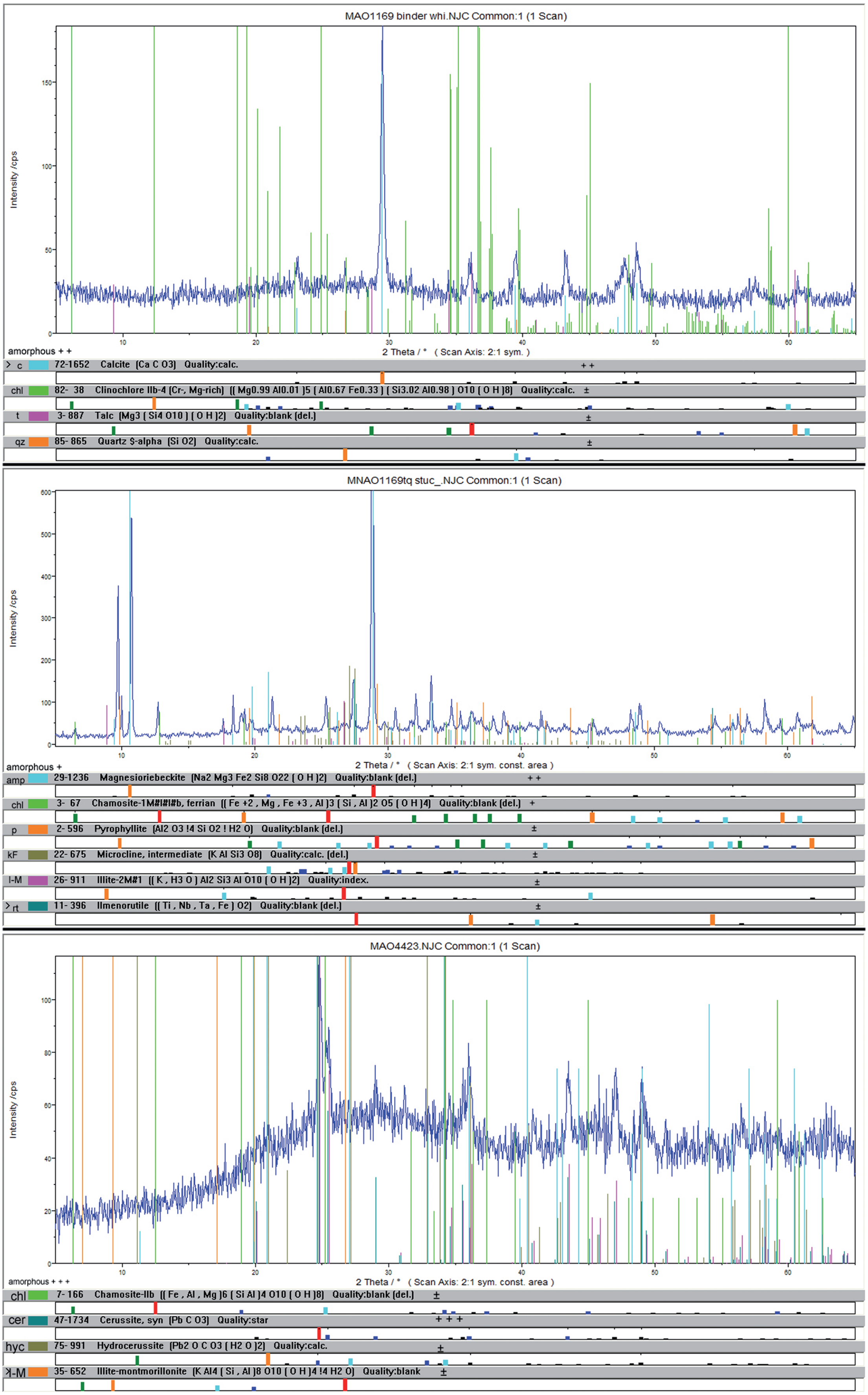


Fig. 6. XRD patterns of the stone (M1169 central) and its residual white binder (M1169 above). Pattern on the bottom refers to sample M4423 from Panr: the white powder reveals a basic lead carbonate

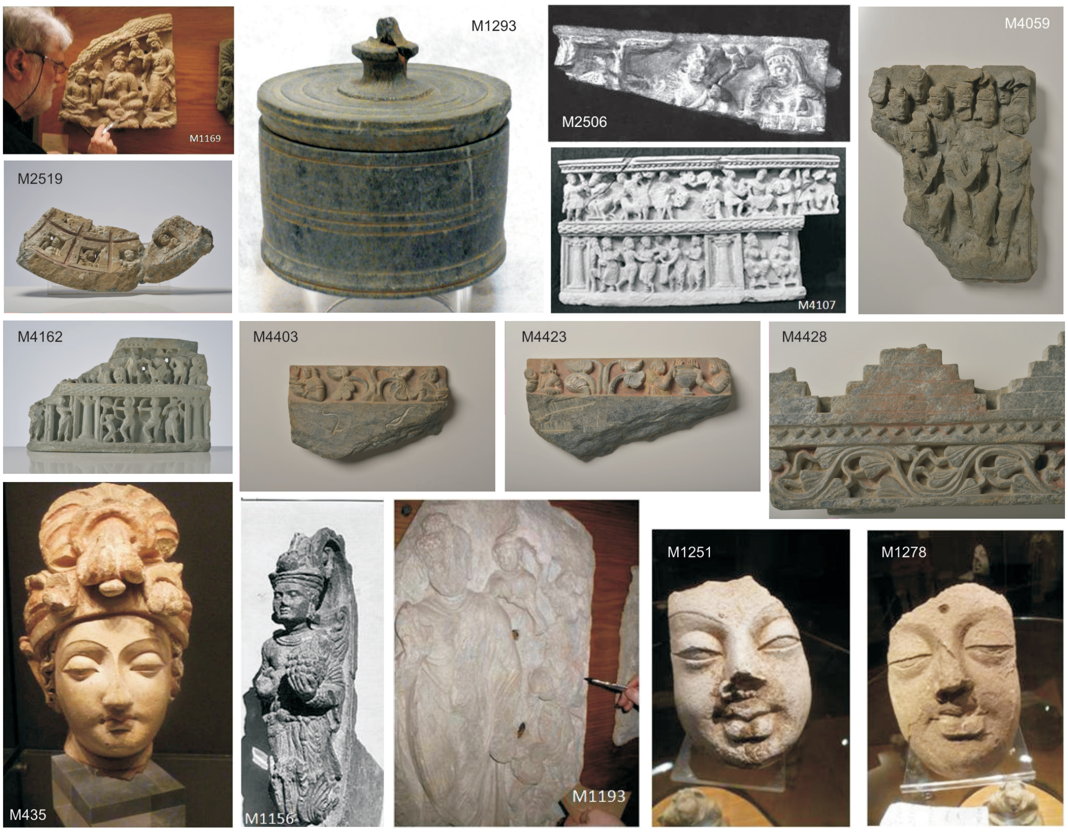
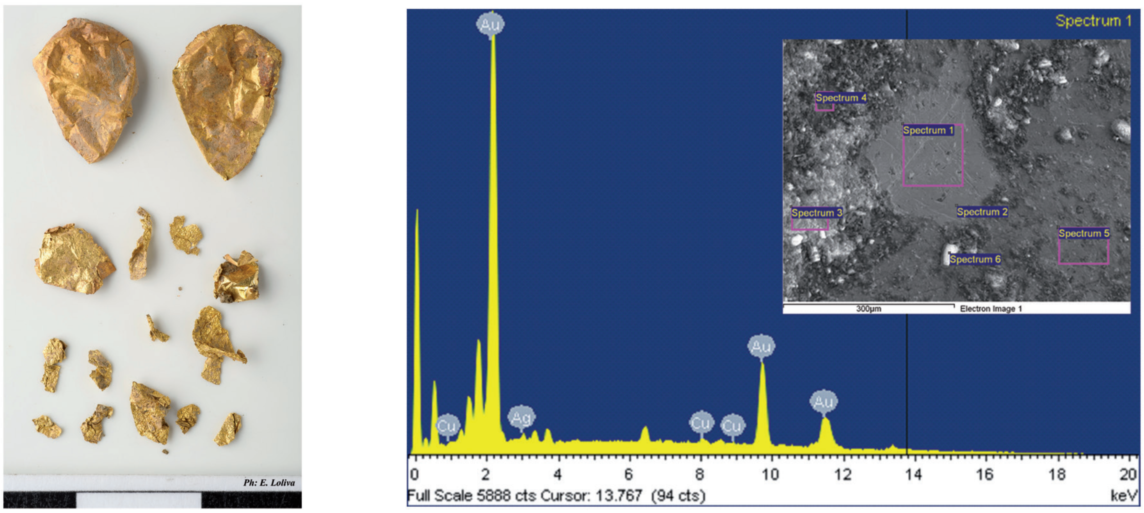


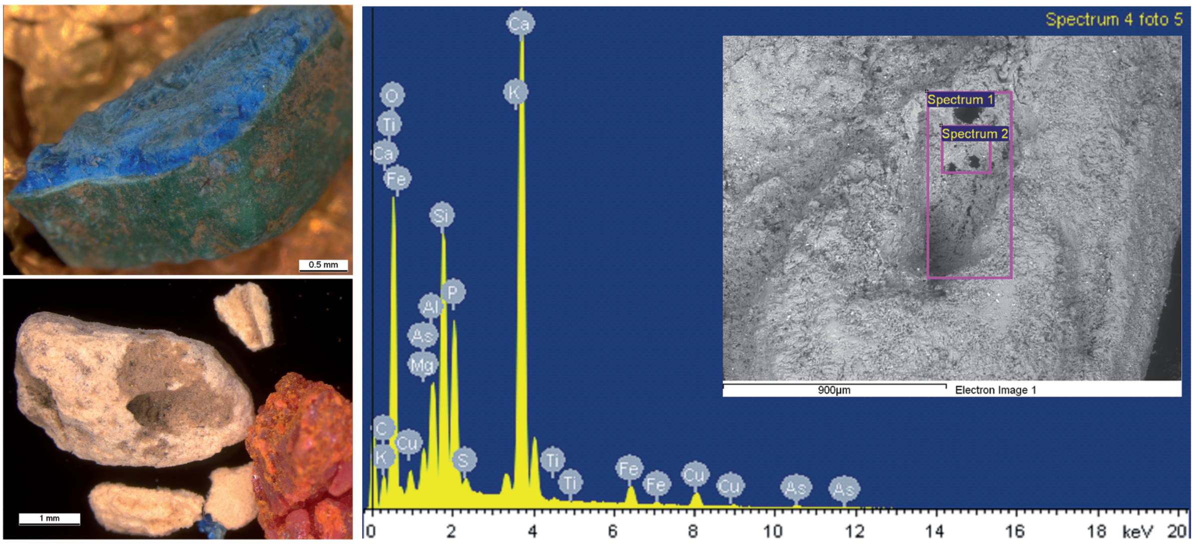
Fig. 7. Examples of artefacts with traces of pigment or preparation layer collected in MNAO: stone (1169, 1293, 2506, 2519, 4059, 4107, 4162, 4403, 4423, 4428) and 'stucco' (435, 1156, 1193, 1251, 1278)



Processing option : All elements analysed (Normalised). All results in weight%

Spectr.	In stats.	C	O	Mg	Al	Si	Cl	K	Ca	Ti	Fe	Cu	Ag	Au	Total
1	Yes											2.63	3.03	94.34	100.00
2	Yes											1.57	2.03	96.40	100.00
3	Yes	6.84	37.44	1.71	5.66	13.25	0.42	2.97	3.38	0.46	10.29	2.14	0.61	14.83	100.00
4	Yes	5.10	36.53	1.39	5.67	14.30	0.41	3.20	2.31	0.47	9.34	2.80	0.93	17.54	100.00
5	Yes	9.70	28.96	1.02	2.51	5.66	0.26	1.01	1.39		3.99	1.77	2.14	41.59	100.00
6	Yes	5.76	37.53	0.94	10.61	14.78		6.12	1.00		3.73	1.02	0.69	17.82	100.00
Max.		9.70	37.53	1.71	10.61	14.78	0.42	6.12	3.38	0.47	10.29	2.80	3.03	96.40	
Min.		5.10	28.96	0.94	2.51	5.66	0.26	1.01	1.00	0.46	3.73	1.02	0.61	14.83	

Fig. 8. Reliquary M1293: "gold leaf" folded in various shapes (e.g. heart) and SEM-EDS analysis



Spectr.	In stats.	C	O	Mg	Al	Si	P	S	K	Ca	Ti	Mn	Fe	Cu	As	Total
1	Yes	12.48	52.01	1.56	3.35	7.94	1.11	0.22	0.91	14.35	0.23	0.28	2.50	2.52	0.55	100.00
2	Yes	13.00	49.88	1.37	4.20	10.05	1.52	0.25	1.20	10.79	0.27	0.17	3.23	3.41	0.66	100.00
3 foto 4	Yes	8.86	51.47	2.08	3.54	7.48	4.23	0.33	0.82	15.38	0.17		2.70	2.31	0.62	100.00
4 foto 5	Yes	9.79	49.76	1.06	2.74	6.21	5.15	0.37	0.73	18.88	0.18		1.85	2.43	0.85	100.00
5 foto 6	Yes	9.81	56.46	2.10	1.12	2.29	1.68	0.29	0.33	23.33	0.14		0.76	1.08	0.59	100.00

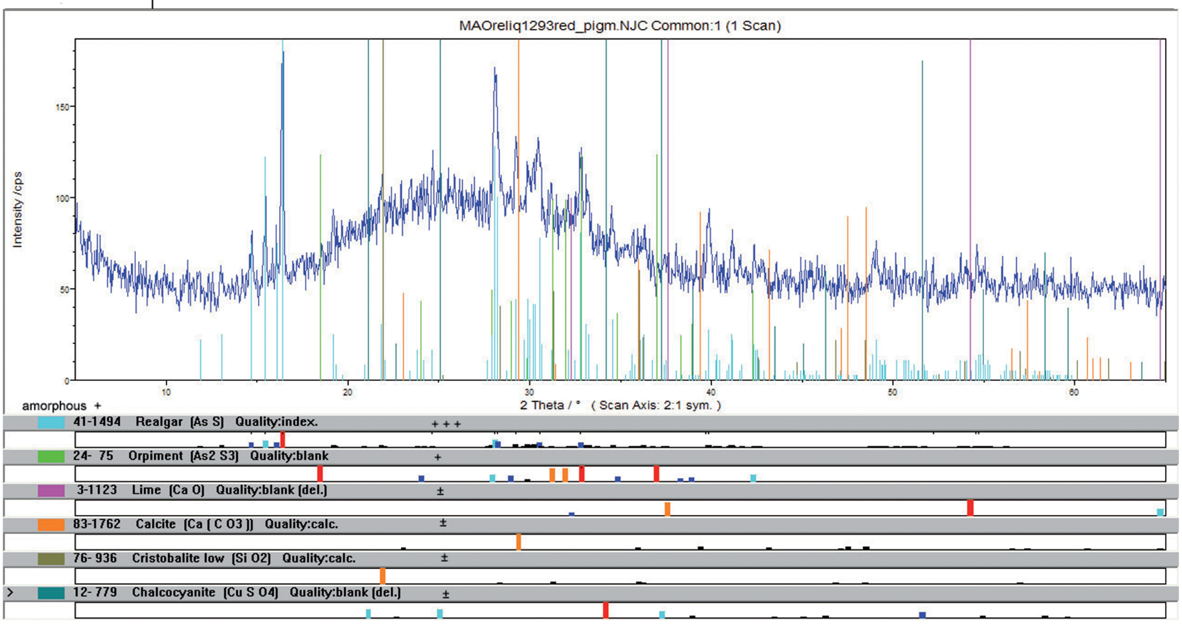


Fig. 9. Grains in reliquary M1293: azurite XX on banded malachite; XRD and SEM spectra of the calcined powder (bones and well crystallized realgar partially altered in orpiment)

of the research by using powder diffraction (XRD) and geochemical analysis on rock samples coming from the Swat. This should help to define chemical correspondence between reliquary and rock and, then, to assign a probable origin of the extraction sites used for the making of the Gandhara statues. It will be fundamental to be able in the near future to collect an exhaustive amount of emerging samples *in situ* and of the possible quarries in the North Western Pakistan area and in Afghanistan.

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