Scientific Research Publishing

Archaeological Discovery, 2023, 11, 107-132
https://www.scirp.org/journal/ad
ISSN Online: 2331-1967
ISSN Print: 2331-1959

Lapis Lazuli Particles on the Turin Shroud: Microscopic Optical Studies and SEM-EDX Analyses

Gérard Lucotte[1], Thierry Thomasset[2]

[1]Institute of Molecular Anthropology, Paris, France
[2]Laboratory of Physico-Chemical Analyses, UST of Compiègne, France
Email: lucottegerard@outlook.fr

How to cite this paper: Lucotte, G., & Thomasset, T. (2023). Lapis Lazuli Particles on the Turin Shroud: Microscopic Optical Studies and SEM-EDX Analyses. *Archaeological Discovery, 11,* 107-132.
https://doi.org/10.4236/ad.2023.112005

Received: January 12, 2023
Accepted: April 1, 2023
Published: April 4, 2023

Abstract

We have studied by optical microscopy and SEM-EDX lapis lazuli particles adhering to a sample of the Turin Shroud. A total number of seventy lapis lazuli particles (and sub-particles) were found on the surface of this sample, and were characterized in details: they are little particles (of between 0.1 and 15 µm of maximal length), of blue colour, and with a spectrum of chemical elements identical to that of the lapis Lazuli mineral. We hypothetise that these particles are residues of painting layers of lapis lazuli that covered initially the Turin Shroud.

Keywords

Turin Shroud, Lapis Lazuli (LL) Particles, Optical and SEM-EDX Studies

1. Introduction

The Turin Shroud (TS) is a well known object in which a body image is imprinted. In 1978, G. Riggi di Numana took some samples of the TS, in particularly corresponding to the Face area, and deposited it on a special sticky-tape; we had access to this sticky tape (cut up in a triangular form) and realized on it preliminary investigations concerning mineral particles (Lucotte, 2012). We have since published some other studies on the triangle, concerning linen fibers (Lucotte, 2015a), pollens and spores (Lucotte, 2015b), red blood cells (Lucotte, 2015c) and hematite, biotite and cinnabar particles (Lucotte et al., 2016).

The mineral LL (Hermann, 1968) contains grains of the cubic lazurite which accounts for the deep blue colour of the ground mineral pigment. As a mineral deposit lazurite is commonly associated with other silicate minerals such as diopside ($CaMgSi_2O_6$), wollastonite ($CaSiO_3$), together with calcite ($CaCO_3$) and iron pyrite. Generally lapis lazuli can be recognized with optical microscopy or under high-magnification scanning electron microscopy and Energy Dispersive X-ray (EDX) analysis. The approximate formula of LL is $(Na, Ca)_8(Al, Si)_{12}O_24S_2$-FeS-$CaCO_3$; that of lazurite is $Na_6Ca_2Al_6Si_6O_{24}[(SO_4), S, Cl(OH)]_2$.

We have already described a little lapis lazuli particle neighbouring the P3 skin debris on the TS triangle sample (Lucotte, 2016). We report now in this study a total number of seventy particles of lapis lazuli found in different areas of the triangle surface.

2. Material and Methods

The material is a small (1.36 mm high, 614 μm wide) sticky-tape triangle (Lucotte, 2017), at the surface of which more than 2500 TS particles were deposited. For practical reasons, the surface of the triangle was subdivided into 19 areas (A to S), the E area being subdivided again in five sub-areas (Ea to Ee).

All the particles described here were studied by optical microscopy and by SEM (Scanning Electron Microscopy)-EDX analysis. Particles of interest were first observed by optical microscopy using a photo-microscope Zeiss, model III 1972. The SEM apparatus used was a Philips XL instrument (an environmental version); GSE (Gaseous Secondary Electrons) and BSE (Bask Scattered Electrons) procedures are used, the last one detecting heavy elements.

Elemental analysis for each particle were realized by EDX, this SEM apparatus being equipped with a Bruker probe AXS-EDX (the system analysis is PGT: Spirit Model, of Princeton Gamma Technology).

Each elemental analysis is given in the form of a spectrum, with kiloelectrons/Volts (ke/V) on the abscissa and elemental peak heights in ordinates. Highly-resolutive (HR) spectras are those where values along the Y axis are enhanced, that permitting a better study of small peaks.

The minimal surface necessary to an EDX analysis is of about 1 μm^2; so, particles which surfaces are below this limit give also EDX analysis of neighbouring particles.

3. Results

The 70 particles (or sub-particles) of LL found are small (of between 0 - 5 μm and 15 μm of maximal length) particles, of various forms (generally with angulous outlines). Their colours are mainly blue or clear-blue; their spectras are those of lapis lazuli particles, but they had also various contents in phosphorous.

Table 1 gives the characterisations of the first twenty-five lapis lazuli particles detected.

There is only one LL particle (a19') in the A area of the triangle. It is a small (1.5 μm of maximal length) particle, of a rectangular form. The colour observed in optic is blue and the spectrum is that of a typical LL particle. The a19' micro-particle is a small scale of lapis lazuli deposited on the a19 particle surface (which is a painting, containing barium sulphate and iron notably).

Table 1. List and characterisations of the first twenty-five lapis lazuli particles.

Numbers	Particles	Areas	Forms	Maximal dimensions	Colours	Spectras	Distinctive features
1	a19’	A	rectangular	1.5 µm	blue	in accordance	scale on a 19
2	b5’	B	squared	3 µm	blue	in accordance	Scale on b5
3	b15	B	ovoid	10 µm	clear-blue	Ca ↗	linked to b16
4	b16	B	ovoid	15 µm	clear-blue	Si, Ca ↗	Linked to b15 and b17
5	b17	B	rectangular	8 µm	dark-blue	Si, Ca, Fe ↗	linked to b16
6	b21’	B	ovoid	6 µm	non-visible	in accordance	near b21; isolated
7	b54’	B	rectangular	2.5 µm	non-visible	Na ↗	near b54; isolated
8	d11	D	ovoid	5 µm	blue	in accordance	near d12; isolated
9	d16	D	triangular	8 µm	non-visible	in accordance	isolated
10	e21	Eb, above	squared	2 µm	blue	in accordance	isolated
11	e9	Eb, below	angulous	4 µm	clear-blue	Si, Ca ↗	linked to e10
12	e10	Eb, below	angulous	4 µm	blue	in accordance	linked to e9
13	e15	Eb, below	triangular	5 µm	blue	in accordance	linked to e10
14	e16	Eb, below	angulous	6 µm	clear-blue	Si, Ca ↗	part of e14
15	e42	Ec, above	angulous	4.5 µm	clear-blue	Si, Al ↗	Stuck up to e41
16	e52	Ec, below	angulous	7 µm	blue	in accordance	linked to e53
17	e53	Ec, below	angulous	9.5 µm	clear-blue	Si, Ca ↗	linked to e52
18	e102	Ee, left	angulous	3.5 µm	blue	in accordance	linked to e102’
19	e102’	Ee, left	elongated	4.5 µm	blue	in accordance	linked to e102
20	e106	Ee, right	triangular	9 µm	clear-blue	Si, Ca ↗	linked to e105, isolated
21	e108	Ee, right	stared	7 µm	clear-blue	Si, Ca ↗	linked to e109
22	e109	Ee, right	squared	4 µm	clear-blue	Si, Ca ↗	linked to e108 and e110
23	e110	Ee, right	triangular	6 µm	clear-blue	Si, Ca ↗	linked to e109 and e112
24	e111	Ee, right	triangular	3 µm	blue	in accordance	linked to e112
25	e112	Ee, right	rectangular	7 µm	blue, with a yellow red center	Fe ↗	linked to e110 and e111

There are six lapis lazuli particles in the B area. Particle b5’ is a square particle of 3 µm of length, of blue colour and with a typical spectrum. The b5’ micro-particle is a small scale of LL deposited on the b5 particle surface (which is a phosphorite).

Particles b15, b16 and b17 are ovoid and rectangular particles of 10 µm, 15 µm and 8 µm of length, respectively. The colour of b15 is clear-blue and its spectrum is relatively rich in silicium and calcium.; the colour of b17 is dark-blue and its spectrum is relatively rich in silicium, calcium and iron (Figure 1 and Figure 2).

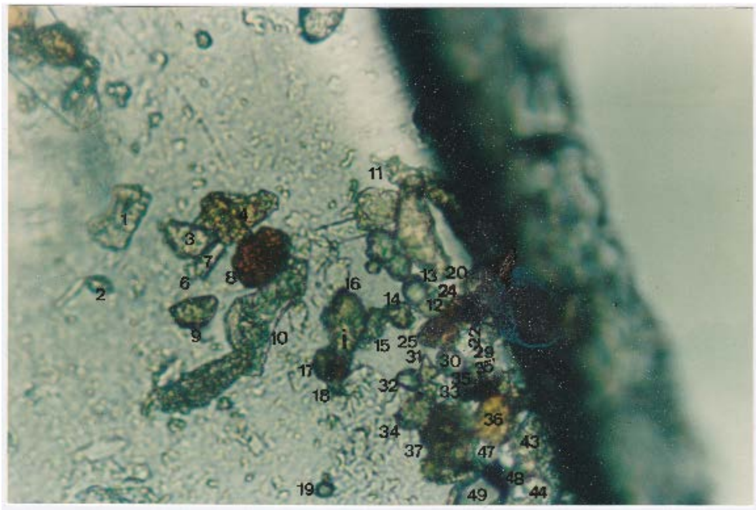

Figure 1. Optical photography (1000×) of the B area of the triangle, showing lapis lazuli particles b15, b16 and b17 (b10 and b11 are linen fibers).

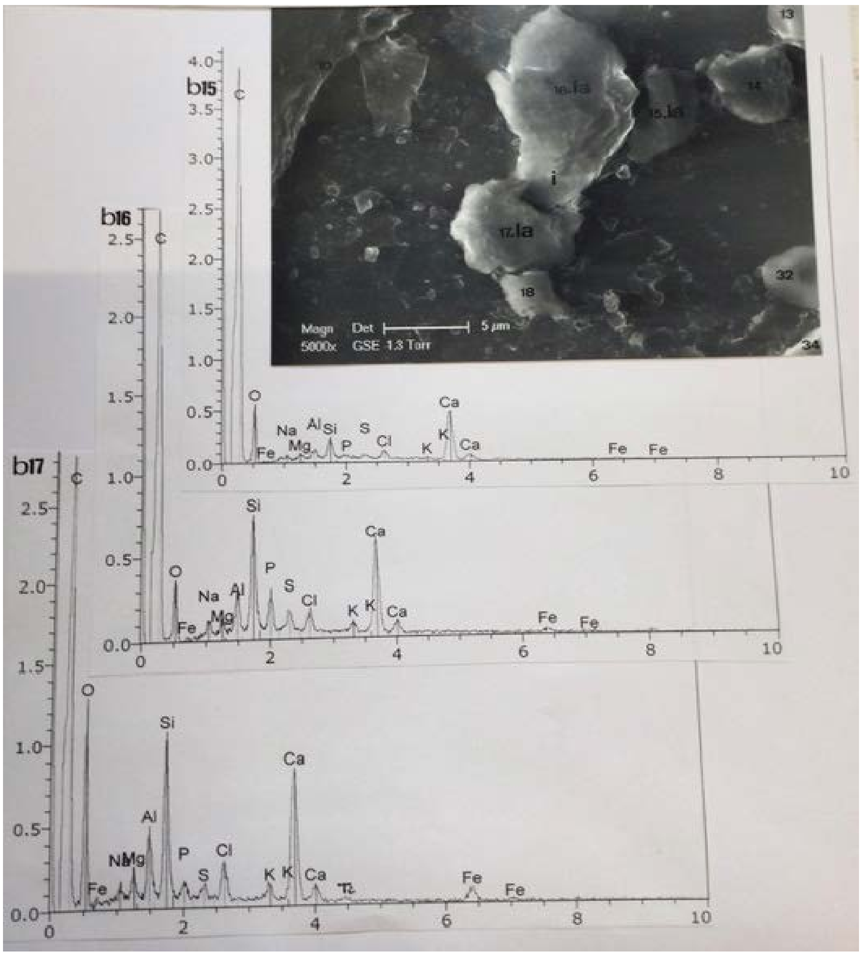

Figure 2. *Above*: SEM photograph (5000×), in GSE, showing lapis lazuli particles b15, b16 and b17 (i: the intermediate part between b16 and b17). *Below*: HR spectras of b15, b16 and b17. C: carbon; O: oxygen; Fe (three peaks): iron; N: sodium; Mg: magnesium; Al: aluminium; Si: silicium; P: phosphorous: S: sulphur; Cl: chlorine; K (two peaks): potassium; Ca (two peaks): calcium; Ti (traces): titanium.

The intermediate (i) region between b16 and b17, which is of yellow colour, is relatively rich in sulphur and iron (**Figure 3**). Particles b15, b16 and b17 are linked together.

Particle b21' is ovoid and of 6 µm of length. The colour is non-visible (because of border effect) and its spectrum is typical. The b21' particle is an isolated form, located near the b21 particle (a ceramic).

Particle b54' is rectangular and of 2.5 µm of length. The colour is also non-visible and its spectrum is relatively rich in sodium. The b54' is isolated and is a micro-scale on the b54 (a pollen) surface (Lucotte, 2015*b*).

There is no lapis lazuli in the C area. Two lapis lazuli particles (d11 and d16) are in the D area. Particle d11 is ovoid and of 5 µm of length; the colour is blue and the spectrum is typical. It is an isolated form, located near the linen fiber d12 (Lucotte, 2015a). Particle d16 is triangular and of 8 µm of length; it is an isolated form. Colour is non-visible (because it is located near the triangular border) and the spectrum is typical.

With sixteen particles (e21, e9, e10, e15, e16, e42, e52, e53, e102, e102', e106, e108, e109, e110, e111 and e112), the E area of the triangle is the richest area in LL particles.

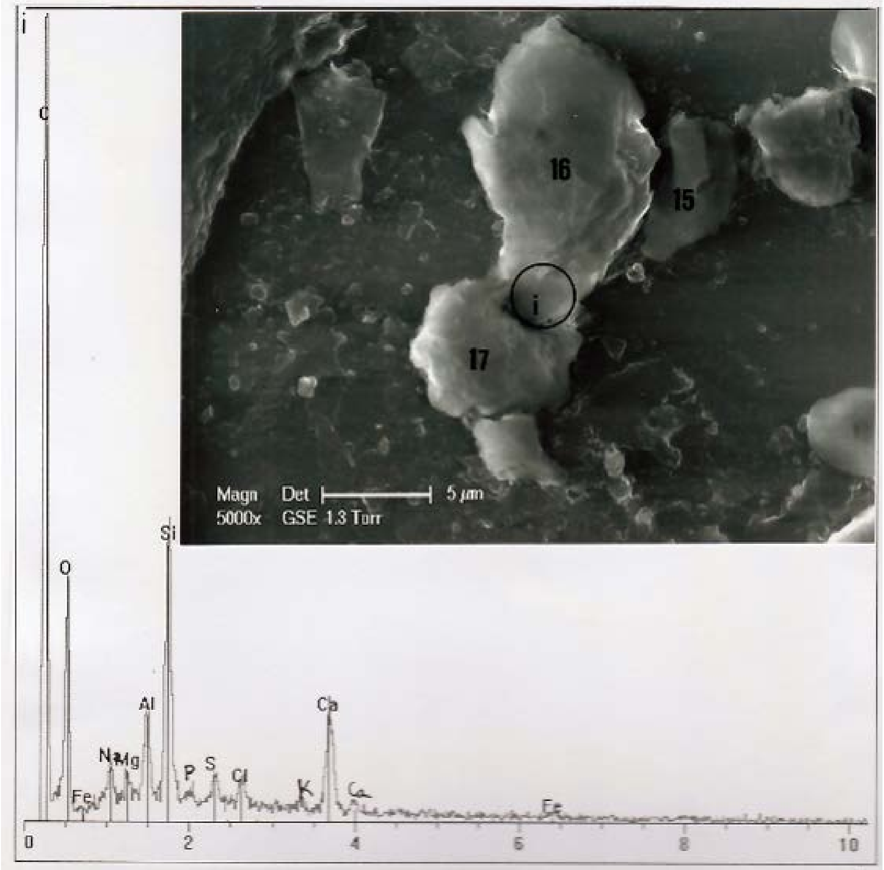

Figure 3. *Above*: SEM photograph (5000×), in GSE, showing particles b15, b16 and b17 (i is the intermediate part between b16 and b17, the circle indicating the surface being studied by EDX). *Below*: HR spectrum in the circle.

The e21 particle (located in Eb, above) is square and of 2 µm; the colour is blue and its spectrum is typical. It is an isolated form.

The e9 particle (located in Eb, below) and the e10 particle (in the same sub-area part) are angulous and of 4 µm of lengths; the colour of e9 is clear-blue and its spectrum is relatively rich in silicium and calcium. The colour of e10 is blue and its spectrum is typical. The e15 particle (also located in Eb, below) is triangular and of 5 µm of length; the colour is blue and its spectrum is typical. The e16 (in the same sub-area part) is angulous and of 6 µm of length; the colour is clear-blue and its spectrum is relatively rich in silicium and calcium. Particle e9 is linked to e10 and particle 15 to e10 also. Particle e16 is some part of e14 (which is a calcium carbonate).

Particle e42 (located in Ec, above) is angulous and of 4.5 µm of length. The colour is clear-blue and its spectrum is relatively rich in silicium and aluminium. The e42 particle is stuck up to e41 (which is an alumino-silicate of potassium, poor in iron).

The particles e52 and e53 (located in Ec, below) are angulous, with lengths of 7 µm and 9.5 µm respectively (**Figure 4**). The colour of e52 is blue and its spectrum is typical; the colour of e53 is clear-blue and its spectrum is relatively rich in silicium and in calcium. The e52 and e53 particles are linked together.

The particles e102 and e102' (located in Ee, left) are two angular and elongated

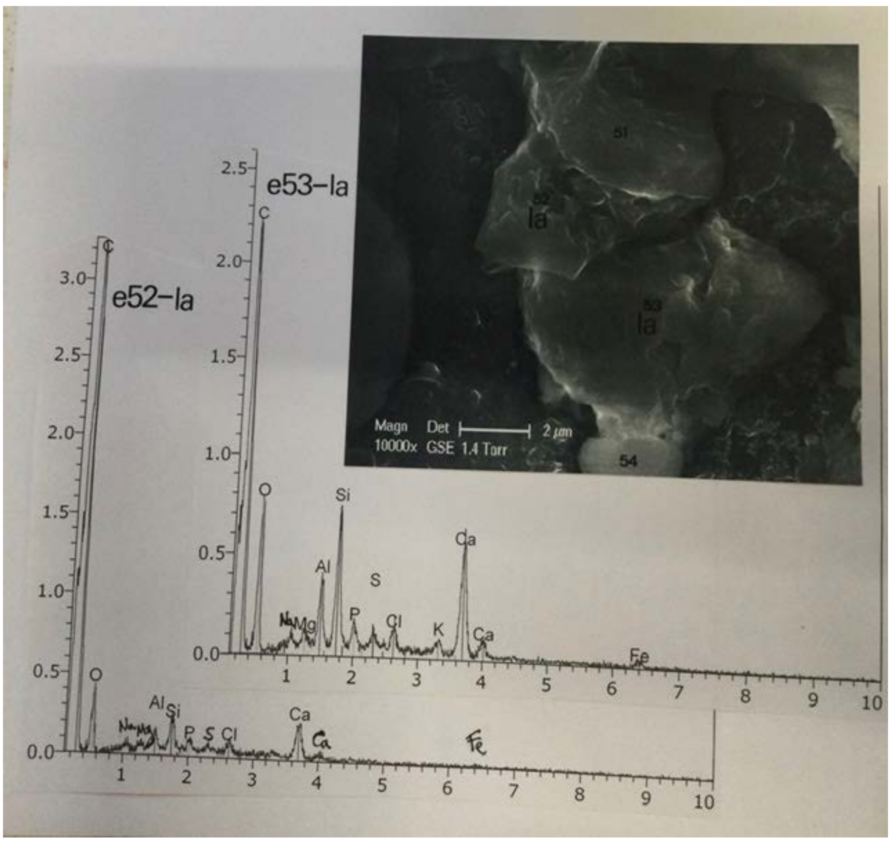

Figure 4. SEM photograph (10,000×), in GSE, showing particles e51, e53 and e54 (e51 is a Diatom and e54 a calcite). *Below*: HR spectras of e52 and e53.

particles, of respectively 3.5 µm and 4.5 µm of lengths. Their colours are blue and their spectras are typical (**Figure 5** and **Figure 6**). The e102 and e102' are linked together.

The particle e106 is triangular and of 9 µm of length (**Figure 5**). The colour is clear-blue and its spectrum is relatively rich in silicium and calcium (**Figure 6**). The e106 particle is located near linen fiber e105 (Lucotte, 2015a).

Particles e108, e109, e110, e111 and e112 (also in Ee, right) are stared, squared, triangular and rectangular, with respectively 7 µm, 4 µm, 6 µm, 3 µm and 7 µm of lengths (**Figure 5**). The colours of e108, e109 and e110 are clear blue and their spectras are relatively rich in silicium and calcium (**Figure 6** and **Figure 7**). The colour of e112 is blue with a yellow-red center and its spectrum is relatively rich in iron (**Figure 7**). The e108, e109, e110, e111 and e112 particles are linked together.

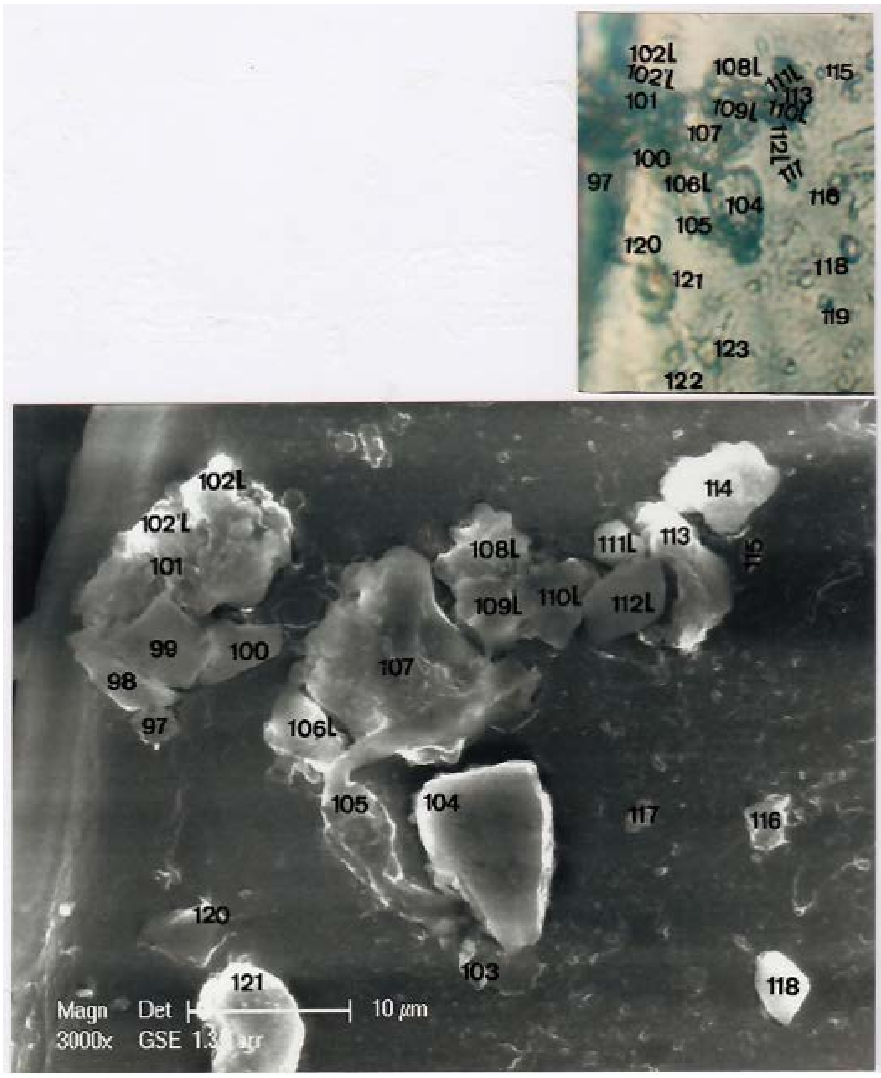

Figure 5. *Above*: optical view (1000×) of the Ec area. Below: SEM photograph (3000×), in GSE, of the Ee area showing the particles e102, e102', e106, e108, e109, e110, e111 and e112.

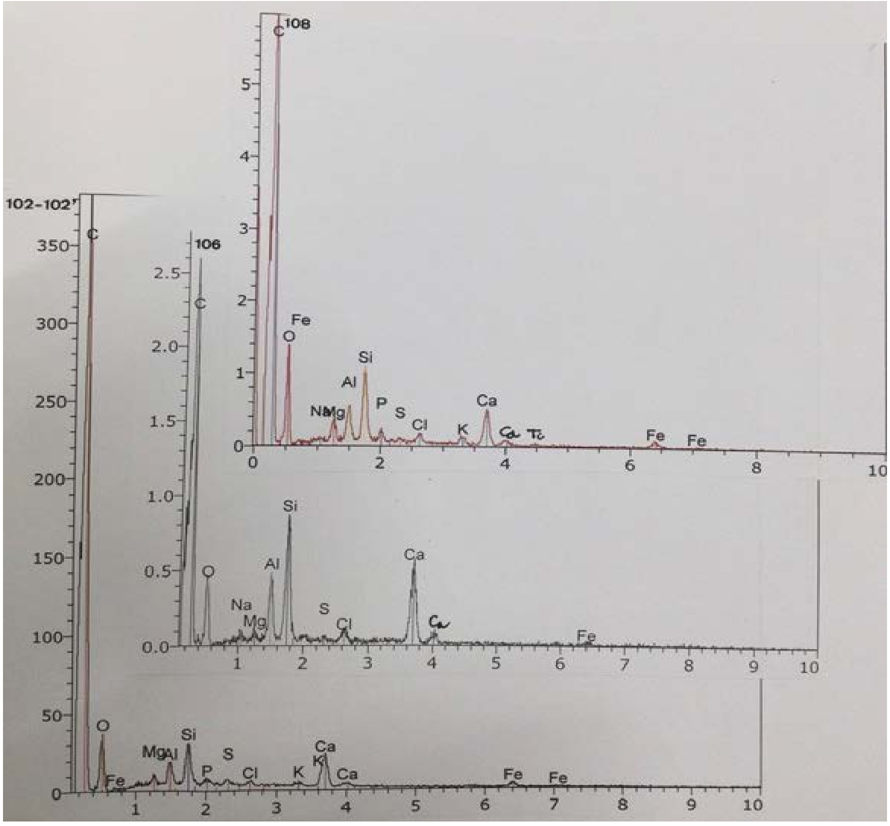

Figure 6. HR Spectras of particles e102-e102', e106 and e108.

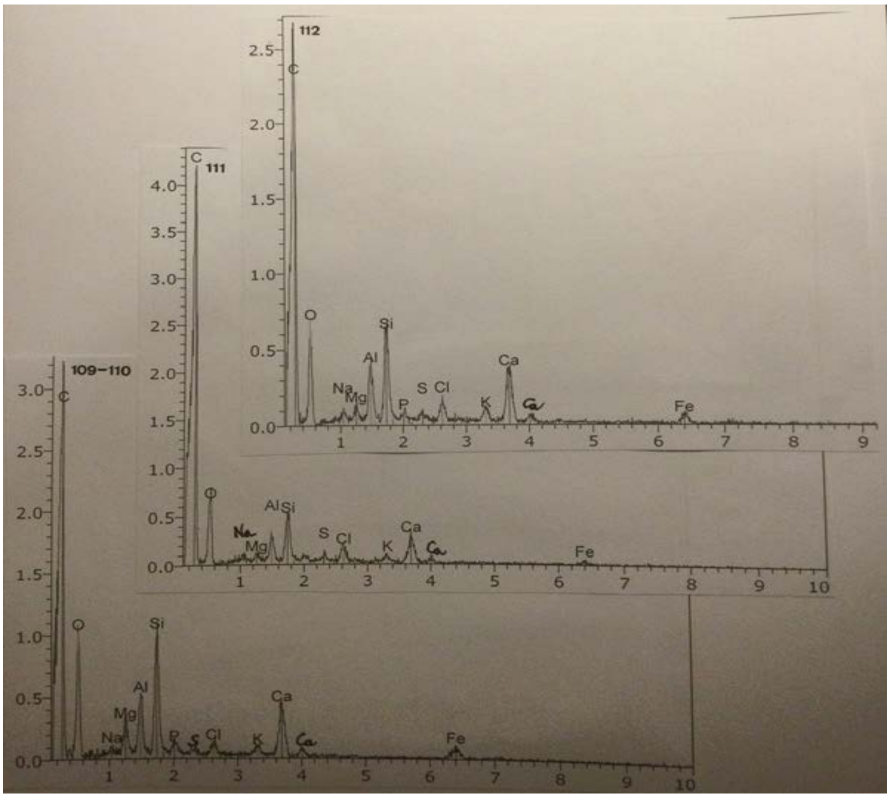

Figure 7. HR Spectras of particles e109-e110, e111 and e112.

The e98, e99 and e100 particles, linked to e101 (a red clay, rich in iron and titanium), have a special morphology (Figure 5 and Figure 8): they are elongated in form, with angulous outlines and their surfaces are smooth. Their colours are intense-blue and their spectras contain copper. They are also lapis lazuli particles, but specially coloured with the blue pigment azurite, of chemical formula: $Cu_3(CO_3)_2(OH)_2$.

There is no lapis lazuli in the F area. Table 2 gives the characterisations of the LL particles numbers 26 to 49.

Particles g72, g74, g76 and g77 (located in the G area) are angulous, triangular, quadrangular and rounded particles, of 6.5 µm, 5 µm, 6 µm and 4 µm of lengths. The colour of g72 is blue (Figure 9) and its spectrum is typical (Figure 10). The colour of g74 is clear-blue, and its spectrum is relatively rich in calcium (Figure 11). The colours of g76 and g77 are clear-blue, and their spectras also relatively rich in calcium (Figure 12). Particles g72, g74, g76 and g77 are lined

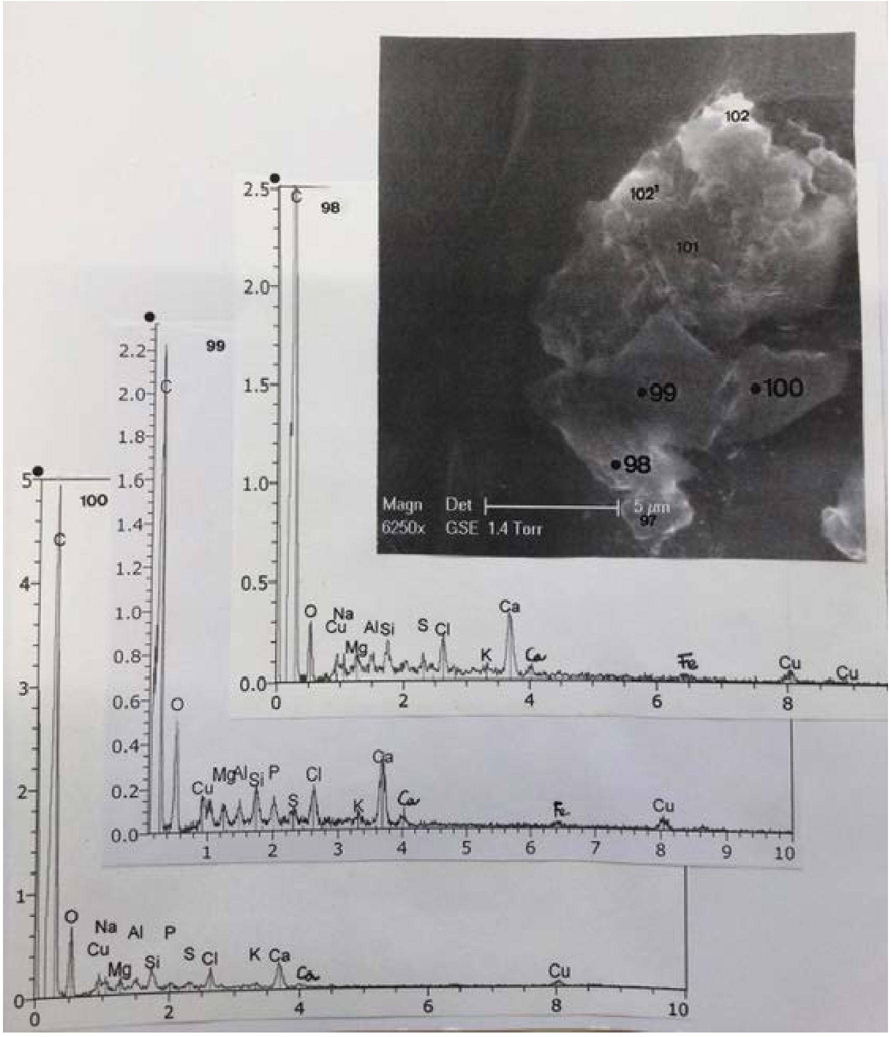

Figure 8. *Above*: SEM photograph (6250×), in GSE, showing particles e98, e99 and e100 (e102 and e102' are lapis lazuli particles; e101 is a montmorillonite; e97 is a calcium carbonate). *Below*: e98, e99 and e100 HR spectras. Cu (three peaks): copper.

Table 2. List and characterizations of the lapis lazuli particles numbers 26 to 49.

Numbers	Particles	Areas	Forms	Maximal dimensions	Colours	Spectras	Distinctive features
26	g72	G	angulous	6.5 µm	blue	in accordance	form of a crystal
27	g74	G	triangular	5 µm	clear-blue	**Ca ↗**	
28	g76	G	quadrangular	6 µm	clear-blue		
29	g77	G	rounded	4 µm	clear-blue		
30	g85	G	angulous	9 µm	blue border, yellow center	**Ca Fe**	isolated
31	h48	H	angulous	7 µm	blue	in accordance	isolated
32	i7’	I	triangular	1.5 µm	blue border	Si ↗	isolated
33	i50	I	rounded	5 µm	blue border	in accordance	isolated
34	i55	I	angulous	3 µm	blue border, white center	Ca ↗	isolated
35	j21-1	J right	angulous	2 µm	clear blue	in accordance	sub-particle on j21
36	j21-3	J right	squared	3 µm	blue border	in accordance	sub-particle on j21
37	j21-4	J right	elongated	4 µm	blue with yellow center	Fe ↗	sub-particle on j21
38	j24-1	J right	angulous	4 µm	clear blue	Si Ca ↗	sub-particle on j24
39	j26	J right	ondulated	3 µm	clear blue	Si Ca	linked to j28
40	j28	J right	angulous	9 µm	black-blue	Fe ↗	linked to j26
41	j28	J right	rounded	0.5 µm	clear-blue	Fe ↗	scale on j28
42	j34	J right	squared	3.5 µm	blue border with white center	Ca Fe ↗	linked to j34’
43	j34’	J right	rounded	1.5 µm	blue border with white center	CaFe ↗	linked to j34
44	j38	J left	angulous	3.5 µm	blue	in accordance	near j38
45	j39	J left	rounded	1.5 µm	blue	in accordance	near j39
46	j40	J left	rounded (bipartite)	3.5 µm	clear blue	Ca ↗	near j39
47	k64	K left	angulous	3 µm	clear-blue	Ca ↗	linked to k66
48	k65	K left	angulous	6.5 µm	blue border, with a yellow center	Ca, Fe ↗	linked to k66
49	k66	K left	angulous	5 µm	blue	in accordance	linked to k64 and k65

up; intermediate particles g72’ (adjacent to g72) and g78 (adjacent to g77) are colourings containing copper.

Particle g85 (also located in the G area) is an isolated LL particle which is angulous and of 9 µm of length. Its colour is with a blue border and a yellow center; its spectrum is relatively rich in calcium and iron.

There is only one lapis lazuli particle in the H area: h48. It is angulous and of 7 µm of length. The colour is blue and the spectrum typical.

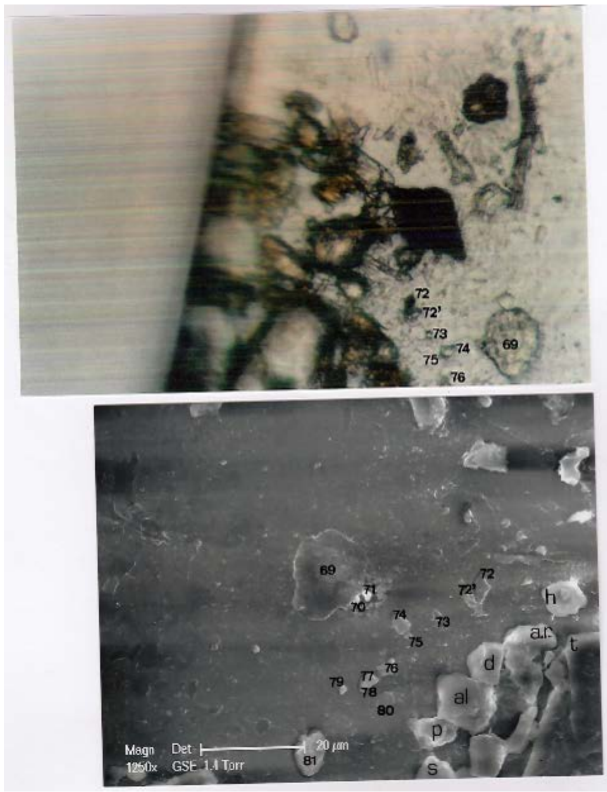

Figure 9. *Above*: inverted optical view (1000×) of the G area, showing particles g72, g72', g73, g74, g75 and g76. *Below*: SEM photograph (1250×), in GSE, of the part of the area G showing particles g72, g72', g73, g74, g75, g76, g77 and g78 (g78' is a colourant, with copper, g73 is a Diatom, g75 is a dolomite; g78 is a colourant; g79 is a ceruse; g80 is a colourant, with copper).

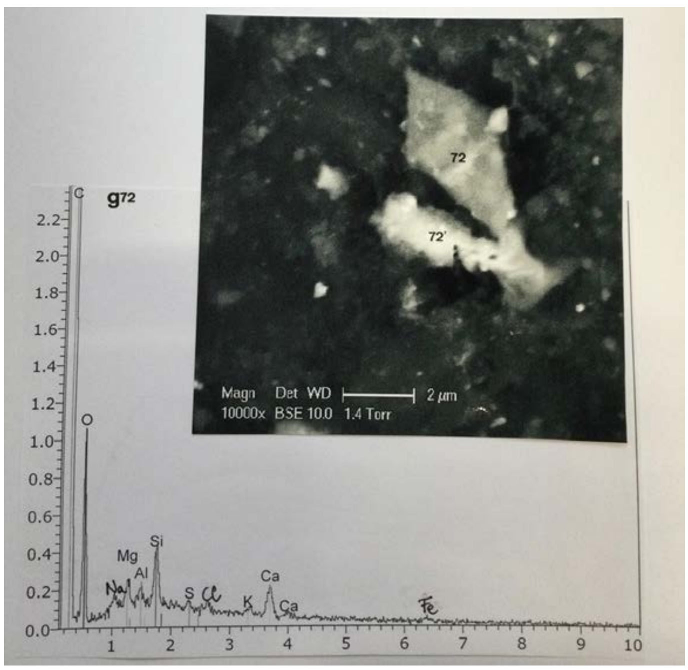

Figure 10. *Above*: SEM photograph (10,000×), in BSE, showing particles g72 and g72'. *Below*: the g72 HR spectrum.

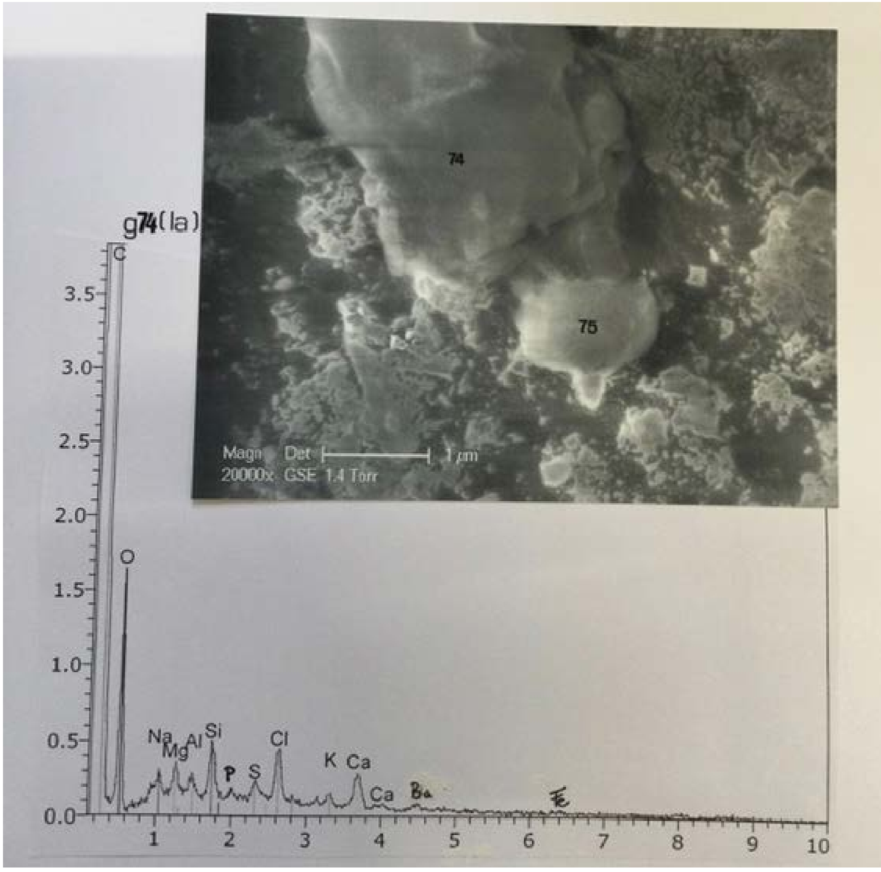

Figure 11. *Above*: SEM photograph (20,000×), in GSE, showing particles g74 and g75. *Below*: the g74 HR spectrum. Ba: barium.

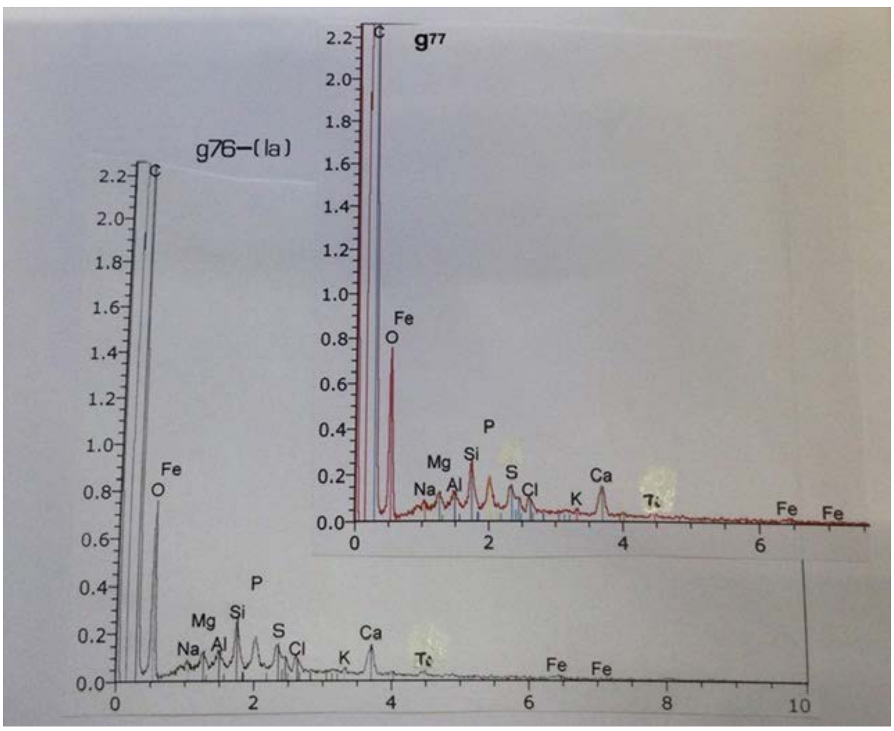

Figure 12. HR Spectras of g76 and g77 particles.

There are three lapis lazuli particles (each are isolated forms) in the I area: i7', i50 and i55. Particle 7' (**Figure 13**) is triangular and of 1.5 µm of length; its colour is with a small blue border and its spectrum relatively rich in silicium. Particle i50 (**Figure 14**) is round and of 5 µm of length, its colour is with a blue border also and its spectrum is typical. The particle i55 is angulous and of 3 µm of length; its colour is also with a blue border but with a white center; its spectrum is relatively rich in calcium.

There are twelve lapis lazuli particles (j21-1, j21-3, j21-4, j24-1, j26, j28, j28', j34, j34' located in J-right, and j39 and j40 (located in J left) in the J area. The j21-1, j21-3 and j21-4 are angulous, squared and elongated, of 2 µm, 3 µm and 4 µm of lengths respectively. Colour of j21-1 is clear-blue and its spectrum is typical. Colour of j21-3 is with a blue border, and its spectrum is also typical. Colour of j21-4 is with a blue border and with a yellow center, and its spectrum is relatively rich in iron. Particles j21-1, j21-3 and j21-4 are sub-particles loaded on the J21 particle (which is a PVC plastic).

The particle j24-1 is angulous and of 4 µm of length. The colour is clear-blue and its spectrum is rich in silicium and in calcium. Particle j24-1 is a sub-particle loaded on the j24 particle (another PVC plastic).

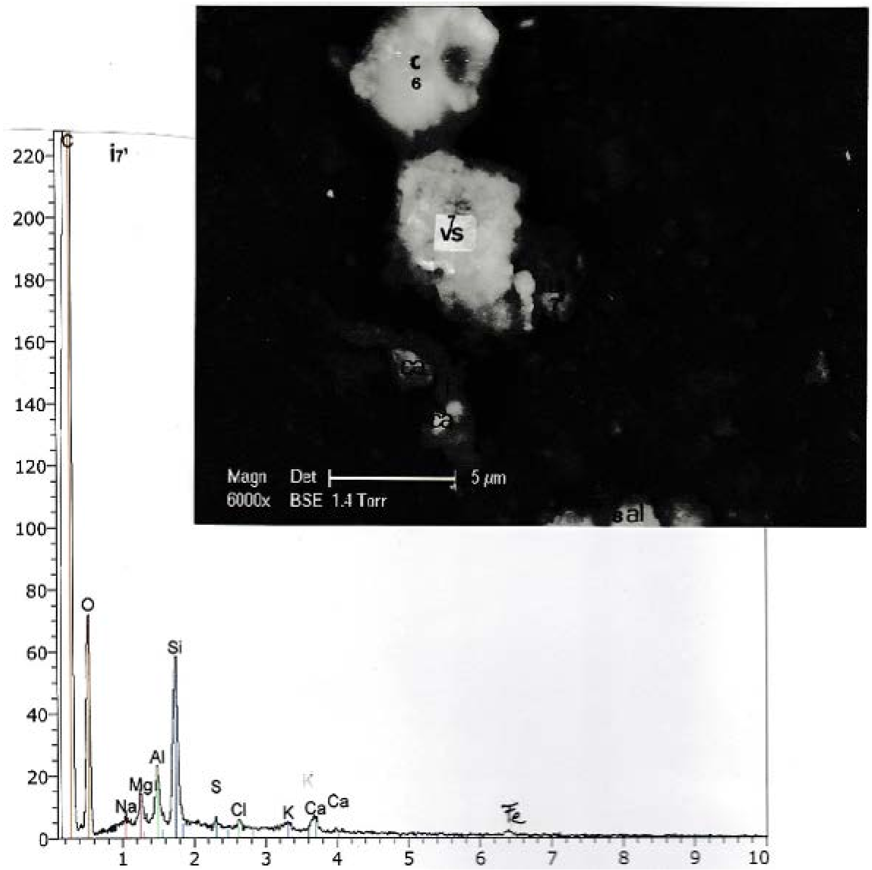

Figure 13. *Above*: SEM photograph (6000×), in BSE, of a part of the I area showing the i7' particle (i6 is a calcium carbonate, i7 is a sodic glass; ca: calcites; f is a fiber; i8 is an alumine. *Below*: the i7' spectrum.

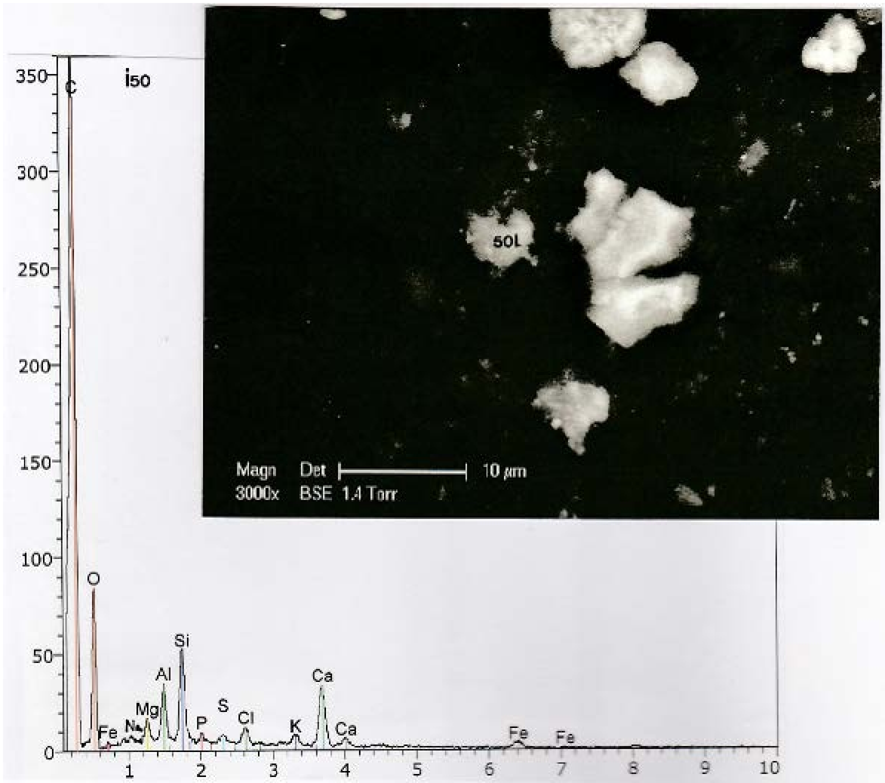

Figure 14. *Above:* SEM photograph (3000×), in BSE, of another part of the I area showing the i50 particle. *Below*: the i50 HR spectrum.

Particles j26, j28 and j28' are ondulated, angulous and round particles, of respectively 3 µm, 9 µm and 0.5 µm of lengths. (Figure 15). The colour of j26 is clear-blue and its spectrum relatively rich in silicium and calcium (Figure 15). The j26 particle is linked to j28. The j28 colour is black-blue and its spectrum is relatively rich in iron (Figure 16). The j28' colour is clear-blue and its spectrum is relatively rich in iron (Figure 16). The j28'particle is a little scale located on the inferior right part of j28.

Particles j34 and j34' are squared and rounded, with respectively 3.5 µm and 1.5 µm of lengths (Figure 17). Colour of j34 is with a blue border and a white center, and its spectrum is relatively rich in calcium and iron. Colour of j34'is clear-blue and its spectrum is also relatively rich in calcium and in iron. The j34 and j34'particles are linked together; at their proximity, the j32 particle is a colouring containing copper.

Particles j38, j39 and j40 (located in J, left) are neighbouring angulous and round particles, of respectively 3 - 5 µm, 1.5 µm and 3.5 µm of lengths (Figure 18). The j 38 and j39 colours are blue and their spectras typical. Colour of j40 (Figure 19) is clear-blue and its spectrum is relatively rich in calcium.

Particles k64, k65 and k66 (located in the K left part) are angulous particles linked together, respectively of 3 µm, 6.5 µm and 5 µm of lengths (Figure 20). The k64 colour is clear-blue and its spectrum relatively rich in calcium. The k64 colour is with a blue border and its spectrum relatively rich in calcium. The k65

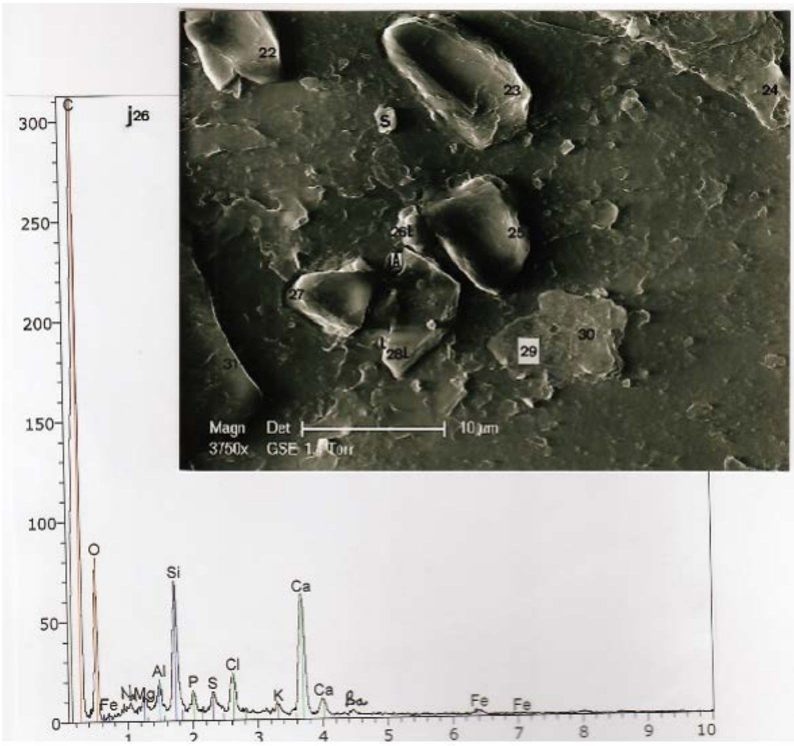

Figure 15. *Above*: SEM photograph (3750×), in GSE, showing the j26, j28 and j28' particles. *Below*: HR spectrum of j26.

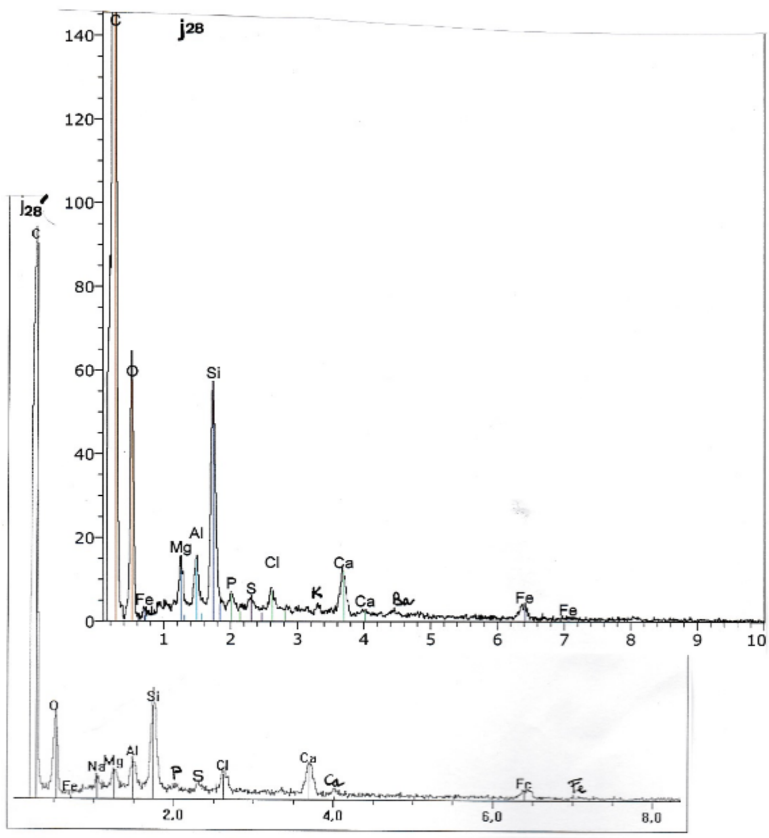

Figure 16. HR spectras of j28 and j28' particles.

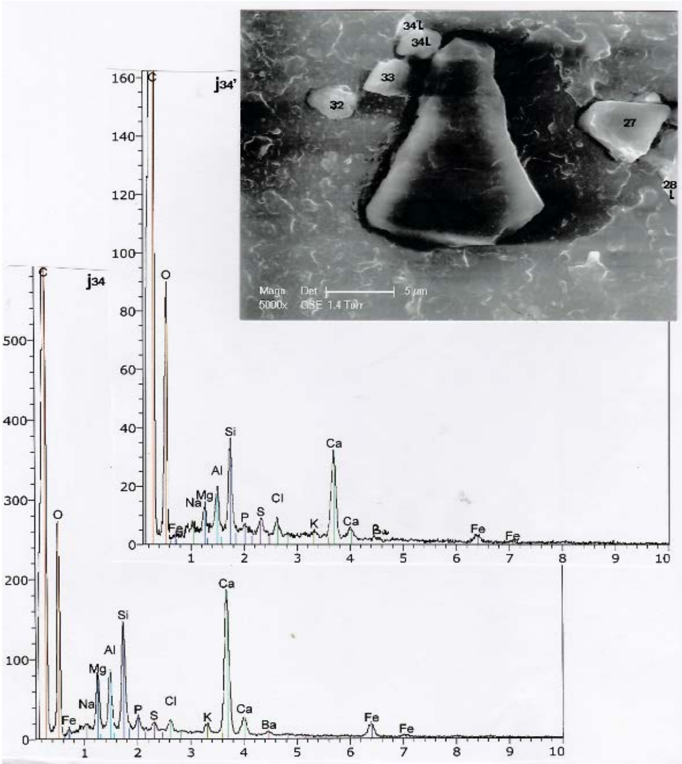

Figure 17. *Above*: SEM photograph (5000×), in GSE, of some part of the J area (J right), showing particles j34 and j34' (j27 is a silice particle; j28 is a lapis lazuli; j32 is a colourant, with copper, j33 is a calcium carbonate). *Below*: HR spectras of j34 and j34' particles.

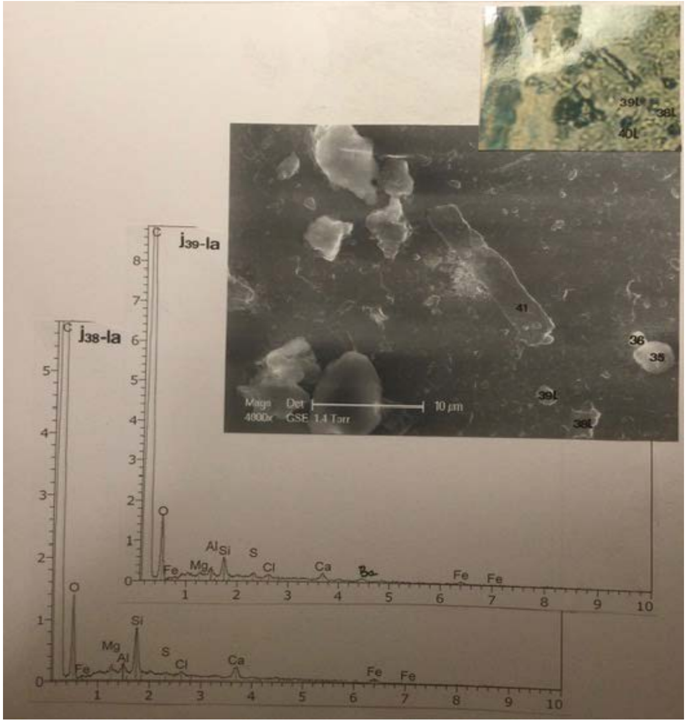

Figure 18. *Above*: optical photograph (1000×), in GSE, of some part of the J area (J left). *Middle*: SEM photograph (1000×) of some part of this I left area showing particles j38 and j39. *Below*: spectras of j38 and j39 particles.

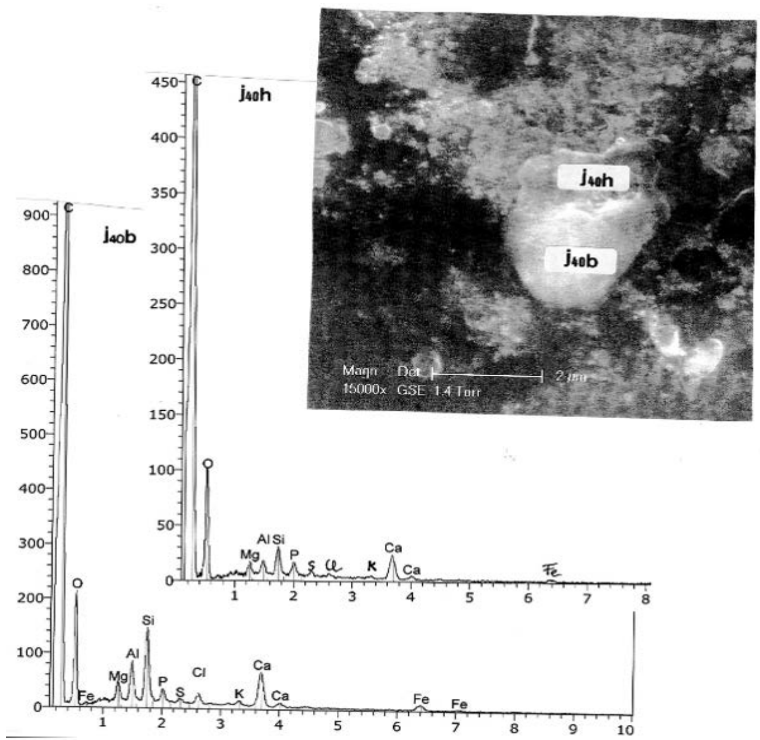

Figure 19. *Above*: SEM photograph (15,000×), in GSE, of the j40 particle (in two parts: j40h and j40b). *Below*: HR spectras of j40h and j40b.

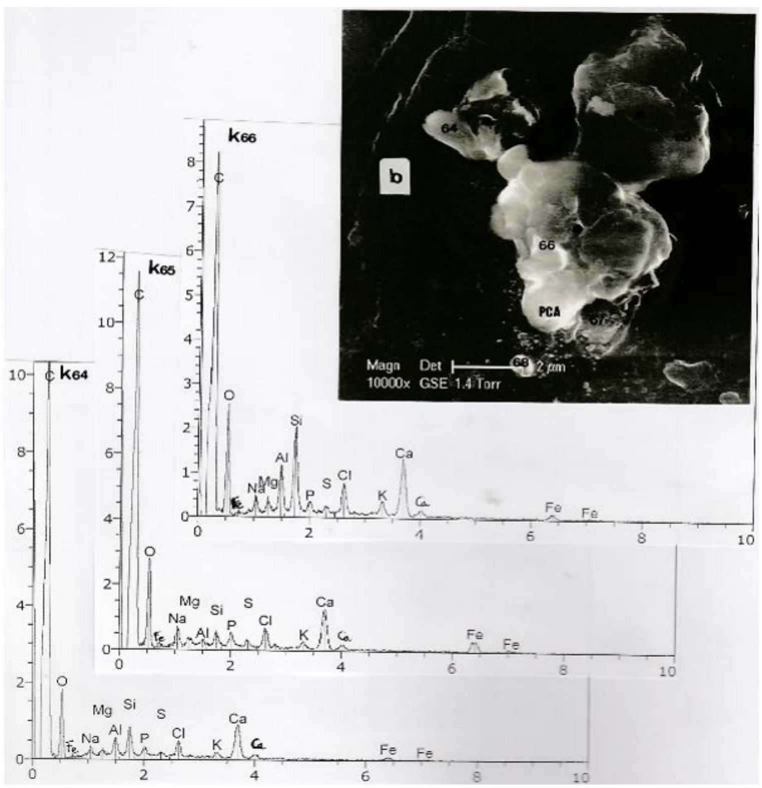

Figure 20. *Above*: SEM photograph (10,000×), in GSE, showing k64, k65 and k66 particles (b is the triangle border of the K area; k67 is a calcium carbonate; k68 is a double carbonate; PCA is a region rich in calcium phosphate. *Below*: spectras of k64, k65 and k66 particles.

colour is with a blue border and a yellow center, and its spectrum is relatively rich in calcium and in iron. The k66 colour is blue and its spectrum is typical.

Table 3 gives the characterisations of the lapis lazuli particles numbers 50 to 70.

Particles l36, l38-15 and l38', located in area L, are angulous and round, and respectively of 2.5 µm, 1 µm and 4 µm of lengths. The l36 colour is clear-blue and its spectrum is relatively rich in calcium. The colours of l38-15 and l38' are blue, and their spectras are typical. Particle l36 is loaded at the superior right side of l38 (a particle of wax); l38-15 is deposited on l38, and l38' is loaded at the inferior left side of l38.

Particle l60 is an isolated particle (located in part L, left) that is triangular and

Table 3. List and characterisations of the lapis lazuli particles numbers 50 to 70.

Numbers	Particles	Areas	Forms	Maximal dimensions	Colous	Spectras	Distinctive Features
50	l36	L	angulous	2.5 µm	clear-blue	Ca ↗	scale on the upper part of e38
51	l38-15	L	angulous	1 µm	blue	in accordance	scale on l 38
52	l38'	L	rounded	4 µm	blue	in accordance	scale on left part of l38
53	l60	L, left	triangular	5 µm	blue border and yellow center	Fe ↗	isolated
54	m31	M	rectangular	1.5 µm	blue	in accordance	isolated
55	m37	M	triangular	4.5 µm	blue-sky	Ca, Fe ↗	isolated
56	n2	N	elongated	5 µm	blue	in accordance	on the same line
57	n4	N	angulous	6 µm	blue	in accordance	on the same line
58	n5	N	rounded	1 µm	blue	in accordance	on the same line
59	n31	N	rounded	3 µm	blue	in accordance	isolated
60	o11	O	angulous	8 µm	clear-blue	Si ↗	linked to o11'
61	o11'	O	rounded	8 µm	clear-blue	Si ↗	linked to o11
62	o20	O	triangular	4 µm	blue	in accordance	isolated
63	p3	P	triangular	3 µm	blue and a white center	Ca ↗	isolated
64	p11	P	elongated	4 µm	blue and a white center	Ca ↗	linked to p12
65	p12	P	elongated	4 µm	blue	in accordance	linked to p11
66	p31	P	squared	5 µm	blue	in accordance	isolated
67	q2	Q	rectangular	6 µm	blue and yellow	Cl, Ca, Fe ↗	near q4a
68	q4a	Q	elongated	6 µm	blue	in accordance	under q4b
69	q4b	Q	rectangular	8 µm	black-blue	in accordance	above q4a
70	s14	S	elongated (multi-part)	15 µm	clear-blue	ClCa ↗	isolated

of 5 μm of length. Colour of l60 is with a blue limit and a yellow center, and its spectrum is relatively rich in iron.

Particles m31 and m37 are two isolated particles located in the M area. They are rectangular and triangular and with respectively 1.5 μm and 4.5 μm of lengths. Colour of m31 is blue and its spectrum is typical. Colour of m37 is blue sky and its spectrum is relatively rich in calcium and iron.

Particles n2, n4 and n5 are three neighbouring particles on the same line located in area N. These particles are elongated, angulous and round, of respectively 5 μm, 6 μm and 1 μm of lengths. Colours of these three particles are blue, and its spectras are typical.

Particle m31 is an isolated and round particle of 3 μm of length. Its colour is blue and its spectrum is typical.

Particles o11 and o11' are two linked particles located in area O. They are angulous and round, and both of 8 μm of length. Their colours are clear-blue and their spectras relatively rich in silicium.

Particle o20, also located in O, is an isolated triangular particle, with a length of 4 μm; the colour is blue and the spectrum is typical.

They are four LL particles in the P area: p3, p11, p12 and p31. Particle p3 is an isolated triangular particle of 3 μm of length; its colour is blue and white and its spectrum is relatively rich in calcium. Particles p11 and p12 are two elongated and linked particles of 4 μm of lengths. The colour of p11 is blue and white and its spectrum is relatively rich in calcium. The colour of p22 is blue and its spectrum is typical.

Figure 21 shows the p5 particle. It is a rectangular particle of about 13 μm of length. Its colour is black-blue and its spectrum is typical of that of a lazurite.

The p31 particle is an isolated and square particle of 5 μm of length. The colour is blue and the spectrum is typical.

There are three lapis lazuli particles in the Q area: q2, q4a and q4b. Particle q2 is rectangular and of 6 μm of length; the colour is with a blue border and an internal yellow micro-ball; its spectrum is relatively rich in chlorine, in calcium and in iron.

Particle q4a (neighbouring q2) is located under q4b. It is an elongated particle of 6 μm of length; the colour is blue and its spectrum typical. The particle q4b is rectangular, of 8 μm of maximal length, and with an intense black-blue colour. Its spectrum is typical.

There is no lapis lazuli in the R area.

The s14 particle is the only lapis lazuli particle found in the S area. It is a voluminous (but truly multipart) elongated particle of about 15 μm of length (Figure 22); the colour is clear-blue and the spectrum is relatively rich in chlorine and calcium.

4. Discussion

Every area of the triangle (but areas C, F and R) contains at least one (particles

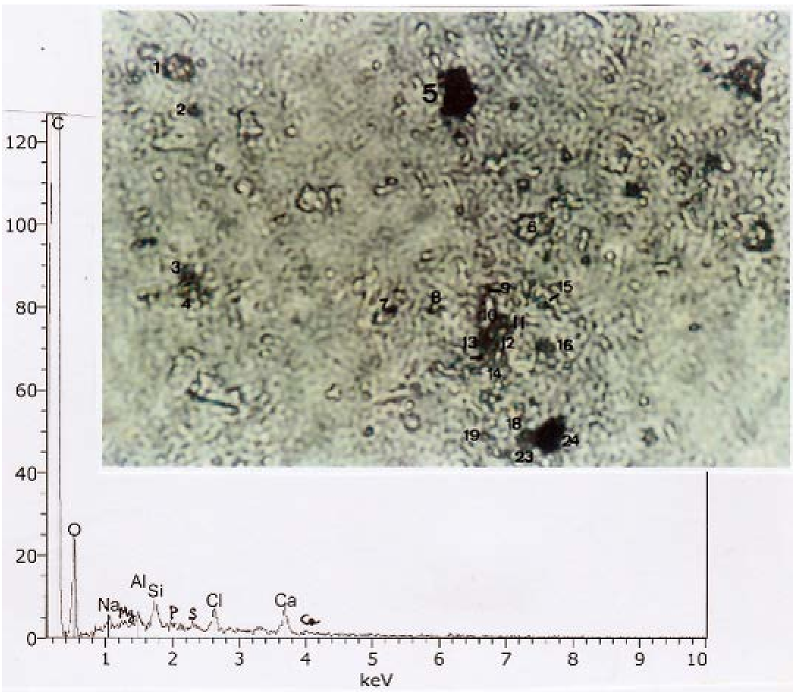

Figure 21. *Above*: optical photograph (1000×) of some part of the P area showing particles p1-p24 (p3, p11 and p12 are lapis lazuli particles). *Below*: HR spectrum of p5 (a lazurite particle).

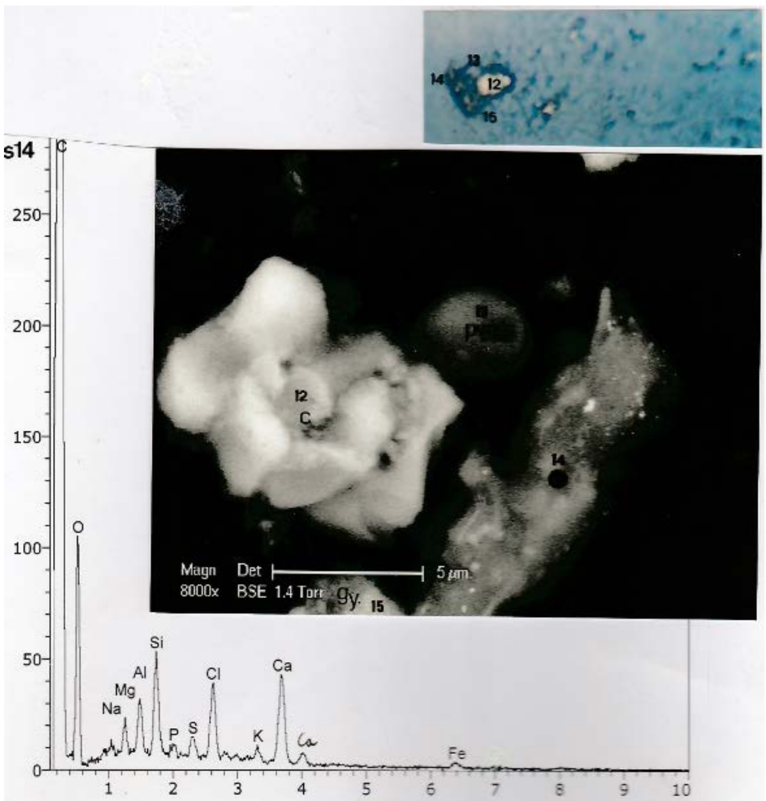

Figure 22. *Above*: inverted optical view (1000×) of some part of the S area showing s12, s13, s14 and s15 particles. *Middle*: SEM photograph (8000×), in BSE, showing the s14 particle (s12 is a Coccolith; s13 is a PVC plastic; s15 is a gypsum). *Below*: HR spectrum of s14.

h48 and s14) lapis lazuli particle. Areas containing the most important numbers of particles are area E (sixteen lapis lazuli particles) and area J (twelve particles). Figure 23 shows the locations on the surface of the triangle of all the seventy lapis lazuli detected: locations of the most important density of lapis lazuli detected (areas E and J) correspond to those richest in particles in general.

The forms of these lapis lazuli particles are generally with angulous outlines, and they are most rarely round. They are little particles, of between 0.5 µm and 15 µm of maximal lengths. Figure 24 shows the distribution of particle sizes (in each class of 2 µm of length); in this diagram the particles of 0.5 to 1.5 µm correspond to lapis lazuli sub-particles (like j28') and those of 10 to 15 µm to composite (like s14) lapis lazuli particles. The modal class of the diagram correspond to particles of between 4 and 5.5 µm of length.

When visible, lapis lazuli particles are generally of blue (or clear-blue) colour; their centers are yellow, red or white.

As a lapis lazuli jewel of reference, we studied the samples of a ring jewel. Figure 25 shows a SEM photograph of three grains of the powder scraped on the surface of this jewel. Although the proportions of the different elements varied between the three samples (Figure 25 and Figure 26), the three main components of lapis lazuli (Si, Al and Mg for the silicate; Ca for the calcite and for other minerals mainly componed of calcium; S and Fe for the pyrite FeS_2) are present

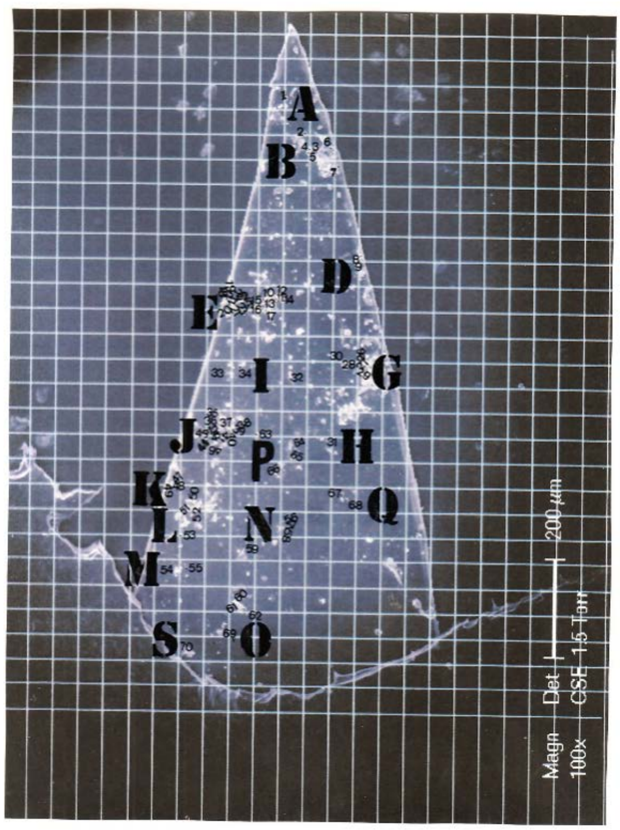

Figure 23. Locations of the 70 lapis lazuli particles detected on the areas concerned of the surface of the triangle.

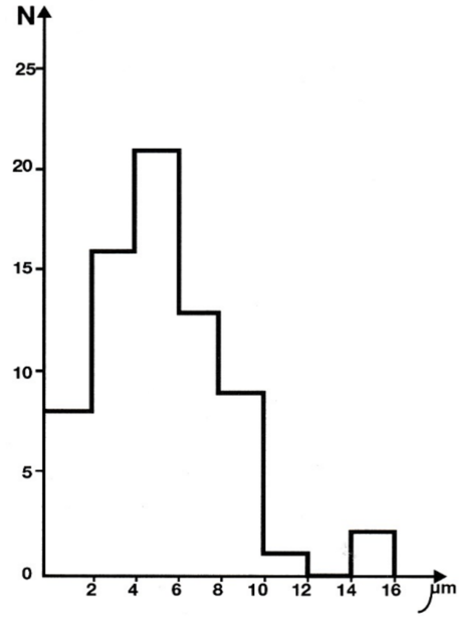

Figure 24. The modal distribution (N: numbers) of the 70 lapis lazuli particles detected among their maximal lengths (in µm).

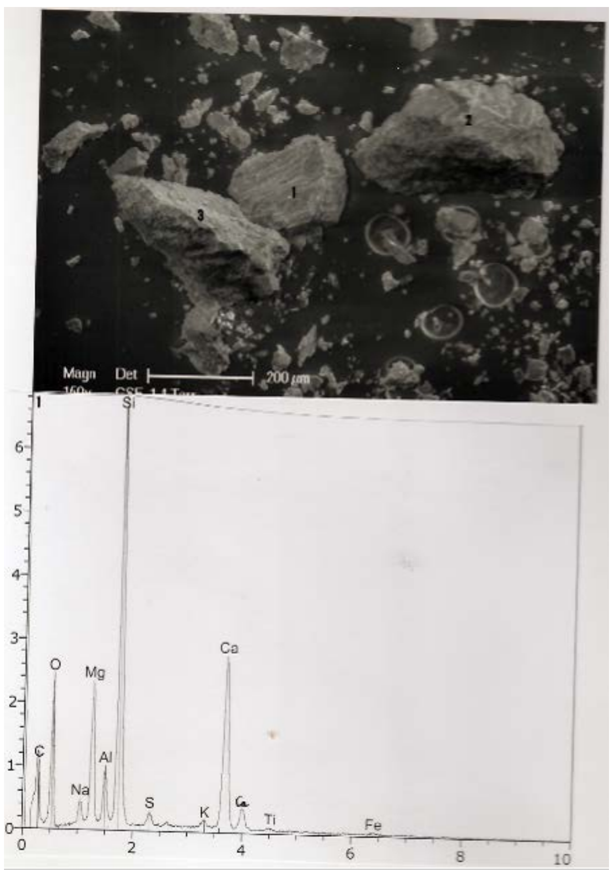

Figure 25. Powder of a ring of lapis lazuli used as reference. *Above*: SEM photograph (150×) in GSE, of three (**1**, **2** and **3**) parts of the powder. *Below*: spectrum of part **1**. C: carbon; O: oxygen; Na: sodium; Mg: magnesium; Al: aluminium; Si: silicium; S: sulphur; K: potassium; Ca (two peaks): calcium; Ti (traces): titanium; Fe (traces): iron.

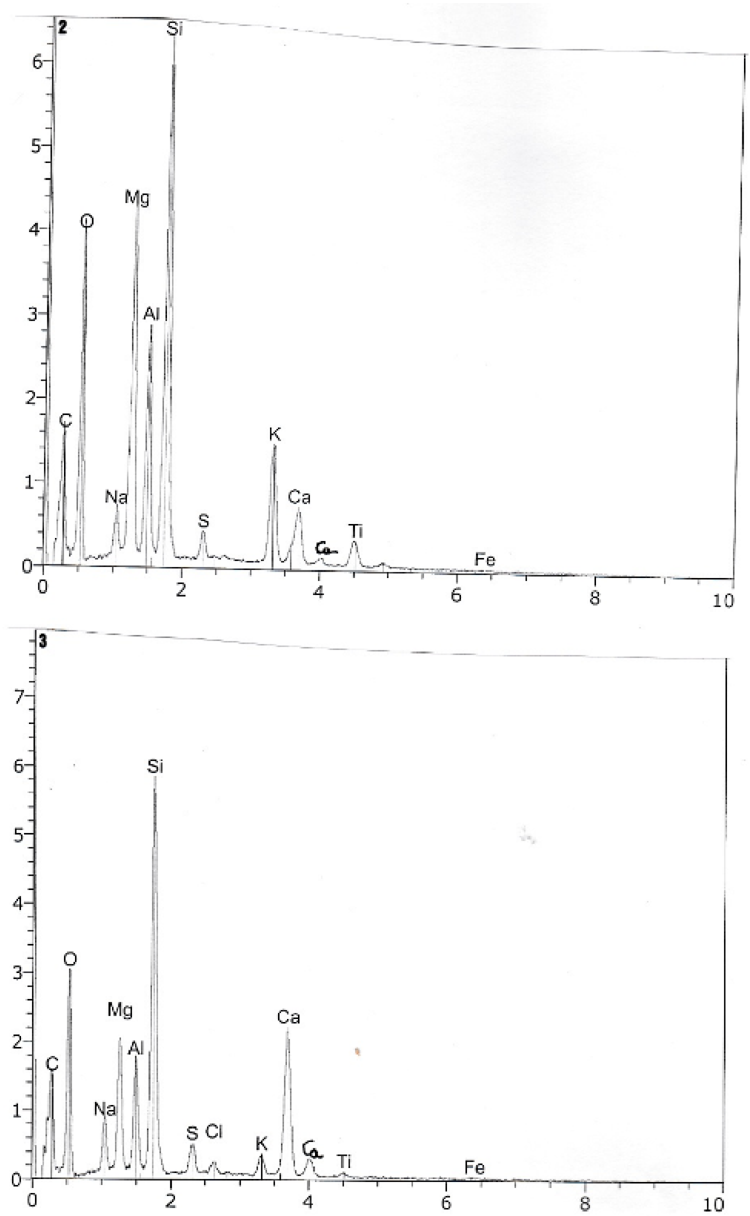

Figure 26. *Above*: spectrum of part **2**. *Below*: spectrum of part **3**.

in the three corresponding spectras.

For our seventy lapis lazuli particles of the TS observed in optical photography, we noted only a weak tendency between the blue colour of the particles with relatively small amount of the calcium element, between the clear-blue colour and relatively elevated level of this element, between the dark-blue colour and relatively elevated values of potassium and sodium, between white in the center of particles and calcium level, and between yellow and red colours in the center and relatively elevated level of iron.

Many lapis lazuli particles of the triangle (like a19', b5', b21', b54', d11, d16, e21, e42, e106, g85, h48, i7', i50, i55 and j26) are isolated, in the sense that they are not closely linked to other particles of the triangle surface. But other particles (e52 and e53, e102 and e102', j28 and j28', j34 and j34', o11 and o11', p11 and p12, and q4a and q4b) are linked in pairs, or in groups of three (b15, b16 and b17; e9, e10 and e15; k64, k65 and k66). The most numerous association between particles is that of the five particles e108, e109, e110, e101 and e112 (**Figure 5**).

But other particles -although separated—are oriented on the same line, as the three particles,n2, n4 and n5; the most numerous case of separated, but oriented particles on the same line, is that the four particles g72, g74, g76 and g77 (**Figure 9**).

These patterns of close associations between particles (or of oriented separate particles on lines) are suggestive of layers of paints formed of lapis lazuli particles on the triangle surface. In that view, isolated particles were residues of these layers covering initially the tissue, after the numerous washings of the Turin Shroud at the time of its long history.

What sort of lapis lazuli mineral pigment is it? Certainly not the artificial ultramarine pigment synthetized in 1828 by J.B. Guimet and that was subsequently adopted as blue by European artists. The approximate formula of ultramarine is Na_{6-10} $Al_6Si_60_{24}S_{2-4}$, and so it does not contain calcium (contrary to lazurite).

Among the lapis lazuli particles of the triangle, we observed a quasi-cubic lazurite particle with a deep blue colour (**Figure 21**). Its spectrum is identic (but Cl) to that of a lazurite mineral of reference (**Figure 27**).

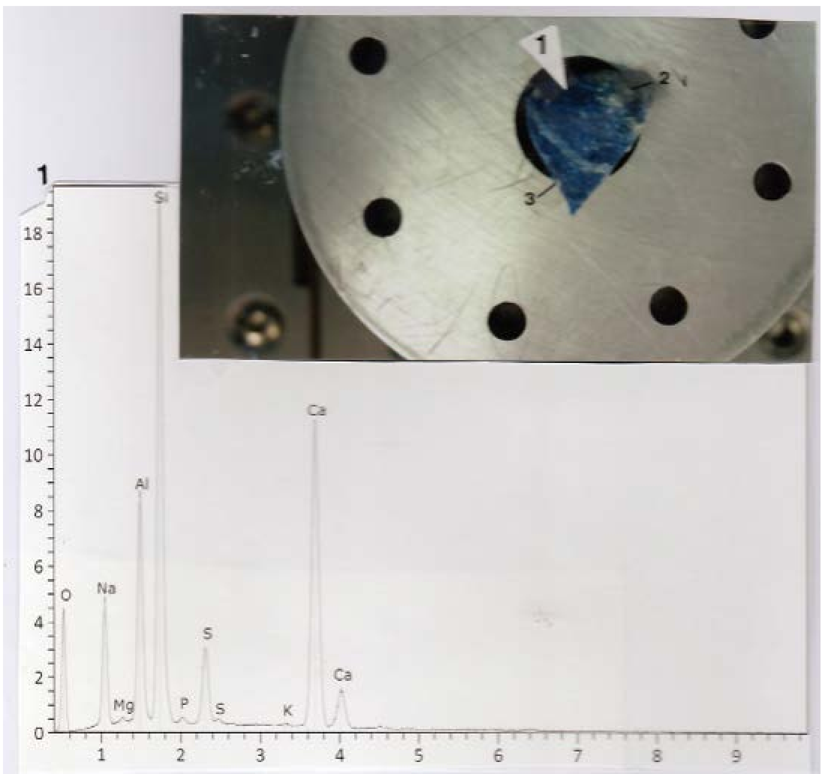

Figure 27. Above: optical photograph (10×) of a lazurite mineral of reference (**1**: the main blue part of the mineral; **2**: a golden point; **3:** a white point). *Below*: the lazurite (part **1**) HR spectrum. O: oxygen; Na: sodium; Mg (traces): magnesium; Al: aluminium; Si: silicium; P: phosphorous; S (two peaks): sulphur; K (traces): potassium; Ca (two peaks): calcium.

We know that the semi-precious stone lapis lazuli was used as glyptic as early as 7000 years ago and for painting starting from the Medieval Age.

Only few sources of lapis lazuli exist in the world, due to the low probability of geological conditions in which it came from. Historical sources of lapis lazuli are in very inaccessible places, such as Afghan and Pamir mountains (Casanova, 2013), and these stones were transported through thousand of kilometres in various trade routes.

Micro-PIXE characterisation of lapis lazuli for a provenance study was realized (Re et al., 2011). The lapis lazuli pigment is common long the Silk Road in wall in wall paint and blue ink for decorative writing (Nöller et al., 2019).

Among particles of the triangle, some lapis lazuli particles are associated with those of azurite (**Figure 8**), which is another blue colouring containing copper; this indicates that, in the Turin Shroud, the lapis lazuli pigment particles were in some case embedded in another less expensive blue material.

In the same order of ideas, we have signalled that intermediate particles g72' and g78 in the lined up g72, g74, g76 and g77 (**Figure 9**), and that the j23 particle near the j34 and j34' associated particles (**Figure 17**) are organic clear blue colourings containing also copper.

5. Conclusion

It is the first time to our knowledge that lapis lazuli minerals are found on the Turin Shroud. Pr Fanti (Fanti & Malfi, 2015) indicated that he found particles of lapis lazuli in the vacuumed dusts of the Shroud, and suspected it as external contaminations that had occurred in the course of centuries.

The seventy lapis lazuli detected on the triangle surface were characterized here for forms and sizes, for colours, and for chemical compositions. They are little particles, of blue colour, which compositions are characteristic of lapis lazuli (with silicate, calcium and pyrite components). Their sizes vary between 0.5 and 15 µm of maximal length, indicating that they are fine particles of a powder. They are present in most areas of the triangle, with a density grossly related to those of other deposited particles in general. The frequently close association observed in numerous of these particles suggests that they are residues of continuous layers of painting.

Why the Turin Shroud was so coloured in blue? The blue of the lapis lazuli (lapis: stone; lazuli: blue) evoke the blue of the sky. Informative for this subject is that the term used for "blue" was the same that for "lapis lazuli" in ancient Egyptian and Akkadian languages (Warburton, 2004). Reasons for the infatuation for the lapis lazuli mineral are due to its profound blue colour and to inclusions in its matter of little gilded fragments that can be assimilated to the starry sky at night.

Acknowledgements

We thank T. Derouin (Natural History Museum of Paris) for his assistance in optical microscopy, and Joelle Salmon for her gift of the lapis lazuli stone of the ring used as reference. The lazurite mineral of reference was purchased from the Carion SA (Paris).

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

References

Casanova, M. (2013). *Le lapis lazuli dans l'Orient ancien.* Editions du Comité des travaux historiques et scientifiques.

Fanti, G., & Malfi, P. (2015). *The Shroud of Turin.* Jenny Stanford Publishing. https://doi.org/10.1201/b18627

Herrmann, G. (1968). Lapis Lazuli: The Early Phases of Its Trade. *IRAQ, 30,* 21-57. https://doi.org/10.2307/4199836

Lucotte, G. (2012). Optical and Chemical Characteristics of the Mineral Particles Found on the Face of the Turin Shroud. *Scientific Research and Essays, 29,* 2545-2553.

Lucotte, G. (2015a). Exploration of the Face of the Turin Shroud. Linen Fibers Studied by SEM Analysis. *International Journal of Latest Research in Science and Technology, 4,* 78-83.

Lucotte, G. (2015b). Exploration of the Face of the Turin Shroud. Pollens Studied by SEM Analysis. *Archaeological Discovery, 3,* 158-178. https://doi.org/10.4236/ad.2015.34014

Lucotte, G. (2015c). Red Blood Cells on the Turin Shroud. *Jacobs Journal of Hematology, 2,* 1-24.

Lucotte, G. (2017). *The Triangle Project* (pp. 5-14). Newsletter 85 of the British Society for the Turin Shroud.

Lucotte, G., Derouin, T., & Thomasset, T. (2016). Hematite, Biotite and Cinnabar on the Face of the Turin Shroud: Microscopy and SEM-EDX Analysis. *Open Journal of Applied Sciences, 6,* 601-605. https://doi.org/10.4236/ojapps.2016.69059

Nöller, R., Feldmann, I., Kasztovsky, Z., Szökefalti-Nagy, Z., & Kovacs, I (2019). Characteristic Features of Lapis Lazuli from Different Provenances, Revised by µXRF, ESEM, PGAA and PIXE. *Journal of Geological Resource and Engineering, 7,* 57-69. https://doi.org/10.17265/2328-2193/2019.02.003

Re, A., Lo Giudice, A., Angelici, D., Calusi, S., Giuntini, L., Massi, M., & Pratesi, G. (2011). Lapis Lazuli Provenance Study by Means of Micro-PIXE. *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms, 269,* 2373-2377. https://doi.org/10.1016/j.nimb.2011.02.070

Warburton, D. A. (2004). Les échanges dans le domaine de la terminologie des couleurs. *Etudes Corses, 59,* 145-168.