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Original article

Do gold particles from the Turin Shroud indicate its presence in the Middle East during the Byzantine Empire?

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ABSTRACT

The following question arose from analysis of dust vacuumed from the Shroud: do these particles show any connection with the gold that could have polluted the linen fabric in past centuries? To find an answer, the composition of gold alloy micro-particles vacuumed at various times from the Turin Shroud are compared with that of gold Byzantine coins minted during the VII–XIII century. The weight percentages of the micro-particles are determined via Energy Dispersive X-ray Fluorescence analysis. The results are arranged in groups to show the similarity in composition between the micro-particles and coins. This process indicated a connection between the micro-particles and the Byzantine coins. In particular, many samples of gold-silver alloy possibly containing copper residual similar to the famous Byzantine Electrum have been detected. It is well known that the Electrum gold-silver alloy is not frequent in the gold coinage of ancient time. The presence of this alloy can be considered typical of the Byzantine Empire, if we exclude the Lydian coins of 6th century B.C. and few other rare cases not applicable for possible correlation with the Shroud. Evaluation of these results, therefore, is compatible with the Shroud's presence in the Byzantine Empire in the period up to 1204 A.D., as many historical clues indicate.

1. Introduction

The Shroud of Turin [1–4], the Holy Shroud or simply the Shroud is an archaeological object as well as a religious object. More research has been performed on the Shroud than on any other ancient artifact in the world. It is, in fact, the only Relic that boasts dozens of publications in specialized scientific journals.

The Shroud is an ancient linen cloth, 4.4 m long and 1.1 m wide, which wrapped the corpse of a tortured man who was scourged, crowned with thorns, crucified and pierced by a spear in the chest. The Christian tradition reports that the Shroud is the burial cloth of Jesus Christ and that he resurrected from within the cloth. Some doctor [5] adds that the corpse was in rigor mortis and that it remained wrapped in the Shroud for not more than forty hours. While the science is not able to surely give a name to this Man, many are convinced that the Shroud is authentic but however, some still think that it is a forgery [6,7].

The double (front and back) body image on the Shroud has been the subject of intense study, but even today, it is not reproducible and cannot be explained scientifically [8,9]. Among the many hypotheses [10,11], that related to a phenomenon acting at a distance like a form of electromagnetic energy appears to be the most acceptable hypothesis currently.

In 1988, the Shroud was radiocarbon dated by three famous laboratories [12]. The resulting date was of 1325 AD with uncertainty of ± 65 years, but this result was widely criticized [13–16] both for procedural and statistical problems. Five independent methods instead assign the first century A.D. as the probable date when the artifact was made [4,17].

On different occasions, dust has been vacuumed from the Shroud and analyzed [18,19]. Many kinds of microscopic material have been identified including organic particles such as fibers, mites, fungi, molds, pollen and parts of plants; inorganic particles like clay, quartz, calcite, etc.; and metal particles like gold, silver, lead and various alloys.

The present study focuses on the particles of gold and its alloys having sizes in the range from 1 to 10 micrometers because they could indicate the Shroud's presence in the Middle East during the Byzantine Empire until it fell in 1204 A.D. [20]. In fact, we believe that fragments of gold coins, jewelry or gold reliquaries in which the Shroud was conserved could have contaminated this most important Relic of Christianity.

The first thing needed is to determine by historical research when the Shroud could have been in contact with gold material before the fall of Constantinople in 1204 A.D.

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2. Shroud history during the first centuries and its possible exposure to gold particles

The weekly exhibition of the Shroud during its stay in Constantinople is attested by the testimony of the crusader knight Robert de Clari, chronicler of the 4th Crusade [21]. In his work, he wrote about the wonders that could be seen before the fall of the city (April 12, 1204): "*Among these there was a church called St. Mary of the Blachernae, where the Shroud (Sydoines) was kept in which Our Lord was wrapped. Every Friday it was elevated all straight, so that it was possible to easily see the image of Our Lord*." [22,23].

Many historians [2,24], by identifying the Shroud as the Mandylion, believe the Relic was in Edessa (currently named Sanliurfa in Turkey), in the early centuries and was in Constantinople until it fell in 1204. Few others [25] think that the Shroud and the Mandylion are two different relics and report that the Mandylion appeared in France where it remained in the Imperial Treasure until 1238 when Baldwin sent it to Louis IX. Nevertheless, they do not specify if the "Mandylion" in question was the original one or a copy.

The authors agree with the hypothesis sustained by many historians for the following reasons:

- like the Shroud, the Mandylion showed not only the face but the full body [26];
- the Mandylion was also called *tetradiplon* which means doubly folded four times [27,28]; when folding the Shroud eight times, only a face is visible like in the description of the Mandylion;
- like the Shroud, the Mandylion showed imprints of sweat and blood [29].

Following this hypothesis, the history of the Mandylion-Shroud indicates various events in which the Relic was exposed to gold particles.

The Mandylion-Shroud or Edessa Image [30] belonged in the first centuries to the Orthodox/Melkite Church. The Codex Vossianus [31] reports that the Mandylion-Shroud was in Mesopotamia of Syria, in the city of Edessa. During festivities, the cloth was pulled out of a golden casket and venerated. In the Doctrine of Addaï [24] (4th–5th century), there is the account of a portrait of Christ being sent to Abgar, king of Edessa. When the king saw the image, he received it with great joy and put it with great honor in one of his palaces.

In the Narratio de Imagine Edessena [26], attributed to Constantine VII Porphyrogenitus, Emperor of Constantinople from 912 to 959, we can read that Abgar gave the order to destroy the statue of a pagan deity that was above the city door and replaced it with the Image of Edessa "fixed to a wooden board and adorned with gold . . .".

Byzantine Emperor Romanus I Lecapenus, in 943, wanted to take possession of the Mandylion-Shroud and sent the army under the command of Armenian General Ioannis Curcuas. Some Islamic sources [32] report both the permanence of the image in Edessa and the bargaining to give it to Byzantium, as well as how it would be transferred into the capital of the Empire. To receive the sacred effigy with great honor, the emperor sent dignitaries bringing candles decorated with gold in their hands. The image was extracted from the reliquary and was venerated with great devotion [26]. The Relic, which contained the precious effigy, came to Constantinople on the evening of August 15, 944, surrounded by a triumphal reception. It was placed in the upper chapel of the church of St. Mary of Blachernae.

Already in Constantinople was a change of casket that was manufactured in the XII century under the Komnenos reign [33]. The reliquary was then transferred to the imperial palace, where it was placed in the chapel of St. Mary of the Pharos, which already hosted many relics of the Passion. The next day, August 16, the emperors revered and kissed the reliquary again and the priests picked it up to bring it into a solemn procession again towards the sea. Then the precious shrine was carried by foot outside the city walls to the Golden Gate. The Relic was then placed on the imperial golden throne in the Chrysotriklinos, an octagonal room. Finally, the Relic was transferred again into the chapel of St. Mary of the Pharos [34].

The image did not have the characteristics of a well-recognizable painting, but of an ethereal imprint [35]. The reliquary that contained the Mandylion-Shroud could have been opened during its long stay in Constantinople. The disclosure of the folded Relic, also called Rakos Tetrádiplon, is confirmed by John Jackson who noticed on the Shroud the existence of folds consistent with the use of a mechanism used to lift the Mandylion-Shroud, so that the entire frontal figure was progressively visible [36]. The existence of machinery and mechanical devices in the city of Constantinople is known [37]. Such a device could have been used to exhibit the Mandylion-Shroud to the faithful.

In the Sacred Imperial Palace, one of the main rooms for exhibition, called the Chrysotriklinos (literally "golden bed"), was specially designed to impress visitors and highlight the emperor's sacredness. Machines were used to lift the throne of the Emperor. The scenery surrounding every emperor's public appearance was made even more impressive by the use of gold covered mechanical devices that were moved by hydraulic pumps. Perhaps such a mechanism was also used to impressively display the Mandylion-Shroud. Wilson [24] argues that a possible exhibition of the Mandylion-Shroud, raised from a reliquary, can be inferred from of the presence of a miniature in a Georgian manuscript of 1054 where it is possible to see a golden drape raised from a reliquary and on it, the Mandylion-Shroud decorated with red crosses.

The historical path of the Shroud from the second half of the XIV century to the present is better known; it was shown in Lirey, France, in 1353, and was in Chambery, France, in 1502. In 1578, it was brought to Turin, Italy, where it remains today. As the Shroud was protected with great care during this time, it is much less probable for the Shroud to have been contaminated with gold particles since the XIV century.

3. The gold Byzantine coins

Constantine the Great introduced the standard gold coin, the *solidus*, in 312 A.D. [20,38–41]. It was made of pure gold 24 carats. Its weight was of about 4.5 g when made in the mint in Constantinople. Its weight and shape were maintained for six centuries and half, up to the reign of Nicephorus II in 963–969. The fineness of the gold solidi remained the same until the reign of Michael IV (1034–1041). Justinian II, just after the Trullan Council introduced the face of Christ on the obverse of the *solidi* that is very similar to the Shroud face [4].

The emperor Nicephorus II (963–969) introduced a new coin, the *histamenon nomisma*, which was thinner and larger than the solidus, during the reign of Basil II (976–1025). Constantine IX in 1042 –1055 transformed the coin into a concave shape. Emperor Alexius I (1081–1118) reformed the monetary system in 1092, which continued up to the fall of Constantinople under the reign of Alexius V in 1204, introducing the golden *hyperpyron* and the *aspron tracky*. The *hyperpyron* was then minted for a century with a fineness greater than reported by Oddy & Niece [1986] of 80–85%, based on the 94% gold measurement for a coin (Ref. 320) of Alexius III.

During the reign of Manuel I in 1143–1180 an economic collapse of the Byzantine Empire produced a debasement of the monetary system and the color of the gold coins became much paler because of the increased content of silver and copper. The gold in this alloy of silver and copper fell to a minimum of only about 30% in weight. The *aspron trachy* instead had initially a fineness of about 30% gradually declining in fineness to about 20% gold by the reign of Isaac II (1185–1195) and to about 0% gold by the fall of Constantinople.

Electrum is an alloy of gold and silver, with trace amounts of copper. Its color ranges from pale to bright yellow. The gold content ranges from 30 to 90%, which increased the profits during the crisis of the Byzantine Empire by issuing currency with a lower gold content than the commonly circulating metal (see the debasement that was evident especially with emperor Manuel I in 1043–1080). This alloy was better for coinage than gold because it was harder and more durable, but widespread trading was hampered by people's evaluation of the intrinsic value of each Electrum coin.

The use of the Electrum begun in the third millennium B.C. in Egypt, but the first metal coins of Electrum date to the end of the 7th century B.C. and were minted in Lydian and East Greek. Even if the use of Electrum for coins is relatively rare in the world, during the debasement, the coins of Electrum alloy were very common in the Byzantine Empire from Romanos Argyros (1028–1034) to Alexius I (1081–1118). Therefore, to find Electrum particles on the Shroud indicates a probable connection with the corresponding Byzantine coin having the same alloy.

Sometimes silver and copper are also contained in native gold and therefore someone could think that these impurities present in the gold coins could derive from this native material. Nevertheless, we must observe that the Byzantine technology was sufficiently advanced to produce pure gold, as we can confirm in the analysis of coins minted up to the XI century.

These results are also important when studying the composition of the contemporary Byzantine jewelry because it has been postulated, even if the hypothesis is not easy to demonstrate, that a possible source of gold for jewelry manufacture would be gold coins. Therefore, the age of a local coinage having the same metal composition is useful to date jewelry of unknown origin [42]. Nevertheless, according to [20], up to about 1000 A.D. gold alloys for jewelry are slightly less fine than the contemporary gold coinage, but from 1000 to 1200 some jewelry was made in a finer alloy than that of the contemporary coinage.

4. Materials and methods

The weight percentages both of golden micro-particles extracted from the Shroud and of golden Byzantine coins are determined via Energy Dispersive X-ray Fluorescence Analysis (XRF-EDS), also called EDX (Energy-dispersive X-ray spectroscopy). The semi-quantitative analysis [43] was performed by means of Eq. (1):

$$Wi = \frac{C_i}{\sum C_n} \tag{1}$$

where C_i is the concentration of the i element and W_i is its relative concentration in percent.

4.1. The dust vacuumed from the Shroud

In 1978 and in 1988 Giovanni Riggi di Numana [18,19], with [44]] vacuumed dust from the backside of the Shroud in the space between the Shroud and the Holland Cloth, and sampled them in various filters named from "*e*" to "*i*", see Fig. 1. Some statistical analyses [45,46] have been done at the microscopic level on this dust.

The 17 golden particles taken from the Shroud dust, which are the object of the present study (see Table 1) were sampled from the filters using carbon sticky tapes mounted on aluminum stubs. These sticky tapes were put in contact with the dust collected in filters "*i*" and "*h*".

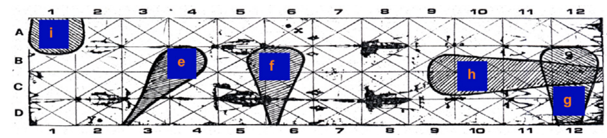

Fig. 1. Areas vacuumed by G. Riggi di Numana with the corresponding filters name.

4.2. Byzantine coins

According to many historians [2,24], the Shroud was in the Middle East between the I and the XIII century A.D. This is the period under analysis. The Crusaders later took it from the Middle East.

If we exclude areas that are not of interest such as America, a numismatic analysis indicates that only the Byzantine Empire used gold alloys like Electrum in this period. Therefore, a possible connection between the alloys of the coins minted both in this period and in this area with the gold dust vacuumed from the Shroud may reinforce the hypothesis of the Shroud being exhibited in the Middle East.

A set of 32 gold Byzantine coins and a medal (see Fig. 2) were washed in soap and water then subjected to EDX analysis. These samples are well recognized in the literature [38,40] and are dated up to 1204 A.D., which is the date for the fall of Constantinople, see Table 2.

4.3. The micro-analysis (XRF-EDS) method

The vacuumed particles were mounted on a bi-adhesive carbon stripe and observed by optical microscopy (Wild 5A stereo microscope) to produce a map of the particles on the stub. This was useful for properly moving on the surface of the sample.

The analysis was carried out with ESEM instrumentation (Environmental Scanning Electron Microscope, FEI mod. Quanta 200) in order to achieve information concerning the morphology and the elemental composition of the particles. With this technique, in the low vacuum mode, it is possible to observe the samples as they are, without any surface preparation or manipulation. Among the types of electron signals generated by the electron beam-sample interaction, secondary electron and backscattered electron signals (BES) were used. BES reflects compositional differences because the grey-level scale of the signal emitted is directly proportional to the mean atomic number (Z) of the sample: the whiter image areas have a higher molecular weight than the darker areas.

The electron microscope is coupled to an XRF-EDS spectrometer (X-Ray Fluorescence- Energy Dispersive Spectrometer by EDAX, Mod. Element) to obtain an elemental analysis having the minimum limit at the element Boron. The spectra acquired in XRF show the peaks of all elements involved, thus allowing a semi-quantitative determination of the composition from the analysis of the net intensities calculated by the peak integral, after subtraction of the background.

Table 1
Shroud particles subjected to the alloy analysis.

Dust golden particle	Alloy
i-24-1-16; f-3.1; i-1.2-25; i-1.24-7; h-24-1-3; h-3-1	Pure gold
19-i–6; 19-i -9	Gold with impurities
19-i-3; 19-i-4; 19-i-5; 19-i-10; 19-i-11; 19-i-fiber-spot 2; 19-i-8.1; 19-i-x; h-6-2	Electrum

Fig. 2. Example of the set of coins and medal tested with EDX.

Table 2
Byzantine coins subjected to the alloy analysis [36,38].

Emperor	Date	Coin Reference
Giustinian II	692–695	1, 315
Basilius II	976–1025	364
Romanus III	1028–1034	47, 124, Cfr. 124
Costantine X	1059–1067	17A, 28
Romanus IV	1068–1071	49, 293
Michael VII	1071–1078	17B, 50, 51, 151, 199, 199A, 295, 296
Niceforus III	1078–1081	134
Alexius I	1181–1118	56, 128, 129
John II	1122–1143	58
Manuel I	1143–1180	28B, 52,127, 222, 224, 405, 407
Alexius III	1195–1197	320, 332
?	600?	Medal 159

All the sample's elements can be analyzed at the same time and their concentrations calculated in weight percent (W%). The spectra are determined in the "spot analyzing mode" for a time interval of 50 s. The results are focused on the metals because the carbon of the background and other impurities have been neglected in the present analysis. We analyzed only the heaviest particles by investigating the bright spots or areas present in the image. We found different types of heavy particles such as lead carbonate, silver sulfate or sulfide, mercury sulfide, and to our surprise a relatively high number of particles of pure gold or gold alloys having sizes variable from 1 to 10 micrometers.

4.4. Possible contamination of gold particles in linen fabrics

The present paper assumes that some gold particles that detached from dresses decorated with gold threads, from rooms and environments decorated with gold and from coins that touched the Shroud could have contaminated the linen fabric. In agreement with Brown and Schweizer [42], it is also assumed that the date of jewelry, dresses or walls that may have scattered their gold dust onto the Shroud can be determined from the date of local coinage if they have the same metal composition.

As it is not obvious that a coin touching a linen fabric could contaminate it with gold particles, the following experiment was performed. A recent gold sovereign coin was rubbed down by a piece of linen fabric very similar to the Shroud. As a result, see Fig. 3, many gold particles having sizes up to about 20 micrometers were detected among the linen fibers.

Fig. 4 shows a gold particle having sizes of about 10 micrometers detected in a fracture of a linen fiber vacuumed from the Shroud (Filter *i*). Therefore, contamination of gold particles can be produced by gold coins rubbing on linen fabric.

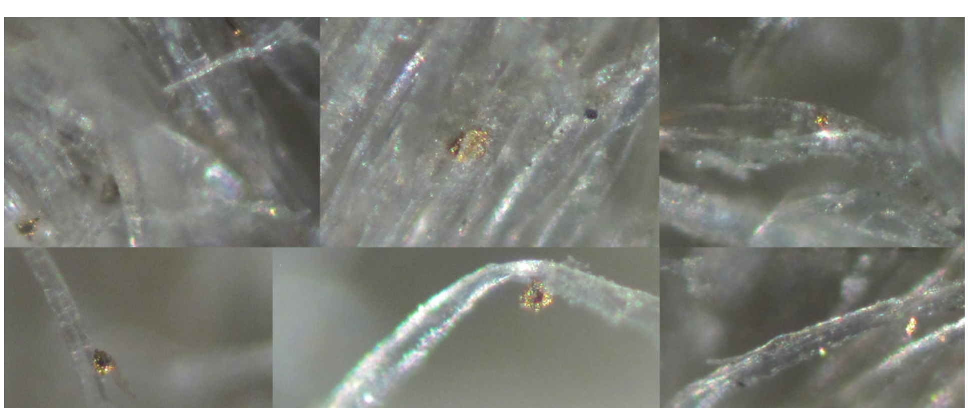

Fig. 3. Example of gold particles dispersed among the linen fibers of a fabric similar to the Shroud. The sizes of the gold particles can be compared to the linen fibers, which have a diameter of 15–20 micrometers.

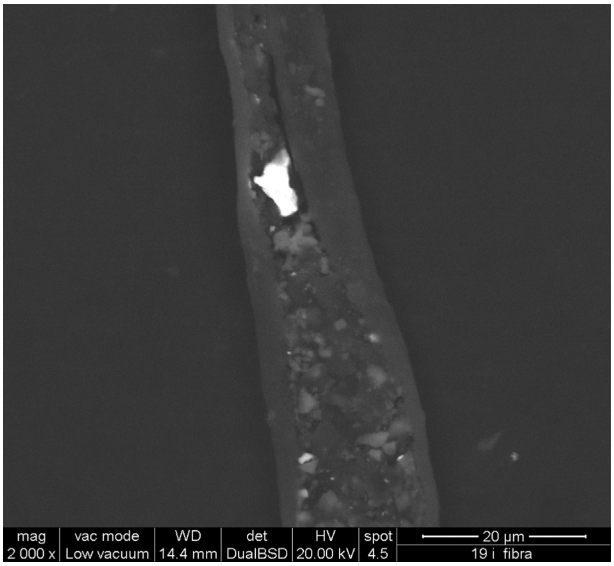

Fig. 4. An example of gold particles of about 10 micrometers detected in a fracture of a linen fiber vacuumed from the Shroud (Particle # 19-i fiber-spot 2).

5. Results

The Byzantine coins and the Shroud particles under analysis have been subdivided into six compatibility-groups on the basis of the different alloy's composition detected by XRF-EDS. Tables 3 and 4 respectively show the percentage of gold, silver, and copper found in the Shroud particles and in the Byzantine coins (*solidi* and *histamena*). The data are approximated to the unit. Their uncertainties roughly vary from ± 1% to ± 2% depending on the indicated values; for example, we can assume 98% ± 2% and 1% ± 1% meaning in this last case a range from 0% to 2%.

We defined the six compatibility-groups of Tables 3 and 4 based on the percentage of gold present in the alloy:

- group 1: 100% of gold;
- group 2: about 95% of gold;
- group 3: about 90% of gold;
- group 4: about 85% of gold;
- group 5: about 70% of gold;
- group 6: about 33% of gold.

For each group, one sample of dust and one coin have been selected and compared in Tables 1SM-6SM of SM (Supplementary Material). They are reported in bold in the first column of Tables 3 and 4 that also report the other non-classified samples with the mark "-".

Due to their small sizes and to the physical process of the generation of X-Ray Fluorescence that considers larger spots than the impact area of the electron beam, the metallic particles from the Shroud show elements not in the metal alloy of coins like silicon, aluminum, calcium, and sodium. To obtain a direct comparison with the coins' alloy, we considered only the elements in the metal alloy like gold (Au), silver (Ag) and copper (Cu) in the calculation of the weight percentages.

The percentages of Au, Ag and Cu are dependent variables, according to the following equation:

$$Au\,[\%] + Ag\,[\%] + Cu\,[\%] = 100\% \tag{2}$$

Table 3 shows that 13 (76%) detected Shroud particles out of 17 (100%) are composed of a gold alloy corresponding to a Byzantine coin under analysis while only 4 (24%) are not. Table 4 shows that 23 (70%) analyzed Byzantine coins or medals out of 33 (100%) are composed of a gold alloy corresponding to a detected Shroud gold particle while only 10 (30%) are not.

Fig. 5A displays the weight percent of the particles from group 1 to group 6 reported in Tables 3 and 4 as a function of the three dependent variables Au [W%], Ag [W%] and Cu [W%]. Fig. 5B shows the weight percent of the particles and coins reported in Tables 1SM-6SM as a function of the two independent variables: Au[W%] and Cu [W%].

Fig. 6A displays the weight percent of gold and silver of all the particles and coins considered in Tables 3 and 4; the dependent variable relative to weight percent of copper (Cu [W%] = 100% − Au [W%] − Ag [W%]) is indicated in the range from 0% to 40% too. It is evident that only some particular combinations of gold-silver-copper are found and that there is a group of Shroud particles of Electrum having a composition of about 80% Au, 10% Ag and 10% Cu that cannot be compared with the coins considered. Future studies could show if this kind of Electrum was perhaps an alloy used in the Byzantine Empire not for the coins but for other objects.

Fig. 6B is the histogram of all the Shroud particles detected in the analysis relative to both gold and silver weight percent. Even if the number of particles is relatively small to perform a reliable statistical analysis, the histogram seems to evidence a bi-modal behavior. One peak corresponds to an alloy of 100% Au and 0% Ag,

Table 3
Percentage of gold, silver, and copper detected in Shroud particles (ordered by gold percentage).

Groups correlated to coins	Sample number	Gold W%	Silver W%	Copper W%	Silver + Copper W%	Kind of alloy
1	i-24-1-16	100	0	0	0	Pure gold
1	f-3.1	100	0	0	0	Pure gold
1	i-1.2-25	100	0	0	0	Pure gold
1	i-1.24-7	100	0	0	0	Pure gold
1	h-24-1-3	100	0	0	0	Pure gold
-	h-3-1 (with silica, red color: glass?)	100	0	0	0	Pure gold
2	19-i-9	96	3	1	4	Gold with impurities
2	19-i-6	93	4	3	7	Gold with impurities
3	h-6-2	89	11	0	11	Electrum
4	19-i-11	85	15	0	15	Electrum
4	19-i-10	85	13	2	15	Electrum
4	19-i-8.1	84	8	8	16	Electrum
-	19-i-x	82	8	10	18	Electrum
-	19-i-4	78	10	12	22	Electrum
-	19-i-3	76	12	12	24	Electrum
5	19-i-5	70	19	11	30	Electrum
6	19-i fiber-spot 2	32	54	14	68	Electrum

The group number of particles correlated with the relative coins is shown in the first column (Group); not correlated particles are indicated with "-".

Table 4
Percentage of gold, silver, and copper detected in Byzantine coins (*solidi and histamena*) well recognized in the literature [36,38] (ordered by age).

Groups correlated to particles	Coin reference	Age [A.D.]	Imperator	Gold W%	Silver W%	Copper W%	Silver + Copper W%	Kind of alloy
1	1	692	Giustinian II	100	0	0	0	Pure gold
1	315	692–695	Giustinian II	100	0	0	0	Pure gold
1	364	976–1025	Basilius II	100	0	0	0	Pure gold
1	Medal 159	600?	?	100	0	0	0	Pure gold
-	124	1028–1034	Romanus III	98	2	0	2	Gold with impurities
2	Cfr. 124	1028–1034	Romanus III	97	3	0	3	Gold with impurities
2	47	1028–1034	Romanus III	94	6	0	6	Gold with impurities
2	17A	1059–1067	Costantine X	96	4	0	4	Gold with impurities
3	28	1059–1067	Costantine X	89	9	2	11	Gold with impurities
3	49	1068 1071	Romanus IV	88	10	2	12	Electrum
3	293	1068–1071	Romanus IV	88	12	0	12	Electrum
2	295	1071–1078	Michael VII	95	4	1	5	Gold with impurities
3	199	1071–1078	Michael VII	92	7	1	8	Gold with impurities
4	51	1071–1078	Michael VII	83	15	2	17	Electrum
5	50	1071–1078	Michael VII	72	26	2	28	Electrum
5	17B	1071–1078	Michael VII	71	25	4	29	Electrum
-	199A	1071–1078	Michael VII	67	30	3	33	Electrum
-	151	1071–1078	Michael VII	66	30	4	34	Electrum
-	296	1071–1078	Michael VII	60	30	10	40	Electrum
-	134	1078–1081	Niceforus III	46	49	5	54	Electrum
-	128	1181–1118	Alexius I	98	1	1	2	Gold with impurities
2	129	1181–1118	Alexius I	95	3	2	5	Gold with impurities
-	56	1181–1118	Alexius I	28	55	17	72	Electrum
-	58	1122–1143	John II	38	62	0	62	Electrum
3	222	1143–1180	Manuel I	89	9	2	11	Electrum
4	28B	1143–1180	Manuel I	87	10	3	13	Electrum
4	407	1143–1180	Manuel I	85	12	3	15	Electrum
-	52	1143–1180	Manuel I	38	41	21	62	Electrum
4	405	1143–1180	Manuel I	87	11	2	13	Electrum
6	224	1143–1180	Manuel I	35	51	14	65	Electrum
6	127	1143–1180	Manuel I	33	62	5	67	Electrum
2	320	1195–1197	Alexius III	94	6	0	6	Gold with impurities
-	332	1195–1197	Alexius III	0	92	8	100	Silver with impurities

The group number of coins correlated with the relative particles is shown in the first column (Group); not correlated coins are indicated with "-".

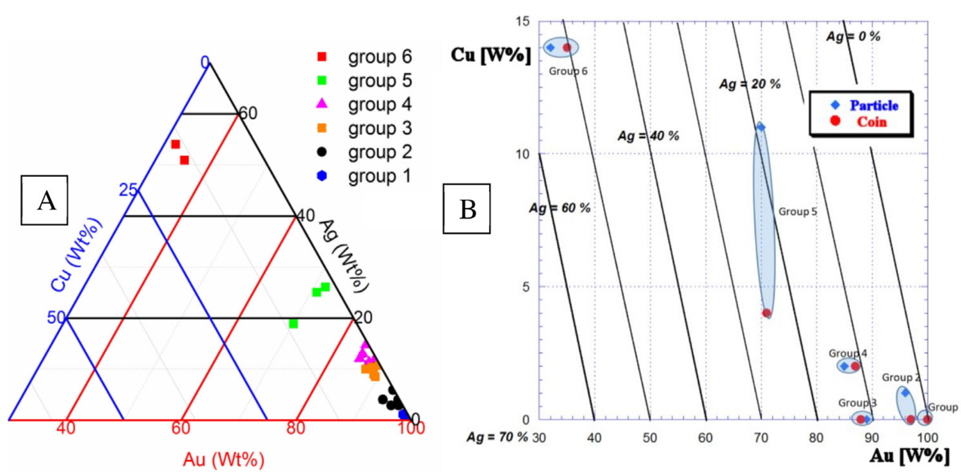

Fig. 5. Two different forms of the plot of weight percent of Au [W%], Ag [W%] and Cu [W%]. Fig. A displays the weight percent of the particles and coins correlated from group 1 to group 6 reported in Tables 3 and 4. Fig. 5B shows the weight percent of the particles and coins reported in Tables 1SM-6SM.

very common for the worked gold of all the ancient times; the other peak, more interesting, corresponds to an alloy of about 85% Au and 10% Ag (with 5% Cu) typical of the Electrum used in the Byzantine Empire.

6. Discussion

Seventeen micro-particles coming from the Shroud, ranging from 0.5 to 10 micrometers, 31 gold and silver Byzantine coins, and a Byzantine gold medal have been analyzed and their compositions in weight percent have been measured with an uncertainty ranging from 1% to 2%. From this data, the micro-particles and coins have been sorted into six groups by their weight percent of gold: 100-95-90-85-70-33% or 24-23-22-20-17-8 carats. Starting from 692 (Justinian II) up to 1197 (Alexius III), the coins taken into consideration belong to the Byzantine Empire especially in the age of the gold debasement that started in 1028–1034 (Romanus III).

Group 1 shows coins and particles made of pure (100%) gold 24 carats. Tables 3 and 4 indicate that there are various particles of this kind on the Shroud and these particles may be due either to pollution produced by coins and jewelry in contact with the Relic before the XI century or due to previous events probably related to those reported in the previous historical section:

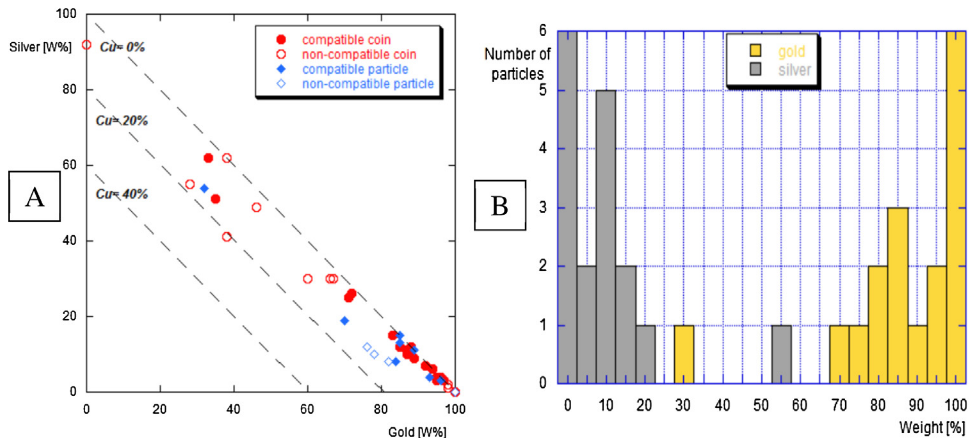

Fig. 6. A displays the weight percent of gold and silver of all the particles and coins considered in Tables 3 and 4; the dependent variable relative to weight percent of copper (Cu [W%] = 100% − Au [W%] − Ag [W%]) is indicated in the range from 0% to 40% too. B is the histogram of all the Shroud particles detected in the analysis relative to both gold and silver weight percent.

Table 5
Gold alloys and corresponding period detected for the groups of Shroud particles and Byzantine coins.

Group #	Gold alloy [%]	Gold alloy [carats]	Shroud particle	Byzantine coin #	Kind and period
1	100	24	i-24-1-16	1	Before the XI Cent.
2	95	23	19-i-9	Cfr. 124	Byzantine gold with impurities 1028-1078
3	90	22	h-6-2	293	Byzantine gold with impurities 1059-1078
4	85	20	19-i-10	405	Byzantine gold debasement-Electrum 1143-1180
5	70	17	19-i-5	17B	Byzantine gold debasement-Electrum 1071-1078
6	33	8	19-i-f.-s.2	224	Byzantine gold debasement-Electrum 1143-1180

- during the first centuries A.D., the cloth was frequently pulled in and out of a golden casket and the Sheet was adorned with gold;
- the Relic was brought in processions by dignitaries bringing candles decorated with gold in their hands and these processions passed under the Golden Gate of Constantinople;
- the Mandylion-Shroud was placed on the imperial golden throne in the Chrysotriklinos. The Relic was shown by raising it from a reliquary probably with the use of a golden mechanical device.

Since pure gold was frequently used in antiquity, the contamination of the Shroud by gold particles during the above-mentioned events is a supposition that should not be neglected.

The groups reported in Table 5 indicate that there are various particles from the Shroud that could derive from direct contact with coins or jewelry made of the same gold alloy. We note that the particles shown in Tables 4SM -6SM are embedded in a fracture of a Shroud linen fiber that was possibly broken by being scraped by a hard object like gold jewelry or a coin.

Among the 17 micro-particles coming from the Shroud, five of them (about 30%) belong to group 1 (24 carats); two of them (about 12%) belong to group 2 (22–23 carats); one of them (about 6%) belongs to group 3 (21 carats); four of them (about 24%) belong to groups 4–5 (17–21 carats); one of them (about 6%) belongs to group 6 (8 carats).

It is interesting to observe that among the 17 micro-particles coming from the Shroud, nine of them (about 53%) are the alloy Electrum. As above-mentioned, these particles do not demonstrate that the Shroud was surely exposed to the pollution produced by the coins or jewelry of the Byzantine Empire, but especially groups from 4 to 6 show a connection of these particles with a particular alloy, the Electrum. It was relatively rare, but typical of the Byzantine Empire during the gold debasement which happened during the XI and XII century; the histogram of Fig. 6A confirms this.

The particles of group 1 (24 carats, Au 100%) could be well coupled with the coins dated from 692 to 1025 A.D. when the history of the Byzantine Empire shows relative economic stability or during the prior period of Edessa. In fact, a numismatic analysis shows that the Byzantine coins minted before the so-called debasement were made of pure gold [36–38].

The particles of groups 2 and 3 show agreement with the gold debasement, especially up to 1180 which are in agreement with Shroud particles of groups from 4 to 6 that reach a majority percentage of silver with the so-called Electrum alloy (8–20 carats, Au 33–85%).

The lowest value of gold (Au 33–35%) is evident in group 6 under the Byzantine Emperor Manuel I (1143–1180) where we find the highest level of silver (Ag 51%) and copper (Cu 14%).

Obviously, the pure gold dust found on the Shroud does not demonstrate its provenance from the Byzantine Empire, although in that period there were jewelry and coins of pure gold. Instead, this dust could be the demonstration of contaminations occurred in other historical periods.

All the results obtained regarding the gold dust have been reported in the paper, but obviously, the most interesting are those related to the Electrum, a typical alloy of the Byzantine era. Notwithstanding this, the paper is not able to surely demonstrate the provenance of the Shroud from the Byzantine Empire, even if the detection on the Shroud of particles of Electrum alloy is a discriminating factor that must not be forgotten in the reconstruction of the Relic's journey.

7. Conclusion

Does the composition of gold alloy micro-particles found in dust vacuumed from the Shroud show any correlation with the gold that could have polluted the linen fabric in the past centuries? To try to answer this question, we compared the gold alloys of the micro-particles vacuumed from the Shroud on different occasions with those of the gold Byzantine coins minted during the period between VIIth and XIIIth centuries. We also have synthesized the historical documents, which report events in which the Shroud could have been contaminated by gold particles. The possibility that the gold micro-particles could have come from golden dresses or other environmental items is reinforced by the fact that some experts associate the gold composition of Byzantine jewelry to the composition of coins in the same period, because they supposed the jewelry was produced by melting coins.

The weight percent of elements in micro-particles and coins have been measured by means of an Energy Dispersive X-ray analysis after proper preparation of the samples. The compositions of 17 micro-particles from the Shroud, 32 gold and silver Byzantine coins, and a Byzantine gold medal have been analyzed and their element weight percent have been measured with an uncertainty ranging from 1% to 2%. From these samples, six groupings of micro-particles and coins have been selected and compared. These groupings are characterized by their gold weight percent: 100-95-90-85-70-33% or 24-23-22-20-17-8 carats.

The coins taken into consideration in our analysis belong to the Byzantine Empire before the fall of Constantinople in 1204 and particularly in the age of the gold debasement begun with Emperor Romanus III (1028–1034). Among the 17 micro-particles coming from the Shroud, five of them are 100% pure gold and could be related to the golden environment in which the Shroud was exhibited before the Byzantine debasement of the XI century.

Two of the micro-particles are composed of gold (93–96%) with metallic impurities of silver and copper and could be related to Byzantine coins struck in the period 1028–1078; four of them are composed of gold (70–89%) and could be related to coins struck in the period 1059–1180; one of them is composed of gold (32%) and could be related to a coin struck in 1143–1180 by Emperor Manuel I.

The histogram of Fig. 6B representing all these Shroud particles seems to evidence a bi-modal behavior. One peak corresponds to an alloy of 100% Au and 0% Ag, very common for the worked gold of all the ancient times. The other peak, more interesting, corresponds to an alloy of about 85% Au and 10% Ag (with 5% Cu) of the Electrum, the gold-silver alloy typical of the Byzantine Empire during the debasement that happened between the XI and XII century; in fact, of the 17 micro-particles coming from the Shroud, nine of them (about 53%) are of Electrum.

These results are compatible with the Shroud being present in the Byzantine Empire prior to 1204 A.D., as many other historical clues indicate.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at http://doi.org.dx/10.1016/j.culher.2019.07.020.

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