Welcome addresses

BENITO RIGHETTI
S&T Attaché – Italian Embassy

As a Scientific Attaché at the Italian Embassy, I am in duty of promoting the bilateral Cupertino between Italy and Hungary in Science and Technology. Today we will see demonstrated how advanced technologies can contribute to enhancement of cultural heritage.

First let me address a warm thank to the very representative authorities who are honouring our meeting: H.E. the Ambassador of Italy P.G. Spinelli, the State Secretary of the Ministry of Cultural Heritage, Mrs Erika Koncz, The Director General of the Hungarian National Museum, Dr. Tibor Kovács who made available for us this wonderful venue.

Let's give a glance to the technologies which will be matter of the symposium: laser beam, X-ray spectroscopy, neutron beam diffraction, ion beam, X-ray digital radiography, ancient DNA analysis, synchrotron light, luminescence, etc. Right a few years ago the use of these technologies was limited to carry on basic research or theoretical investigations. A remarkable step has been made in a few years toward the employment of these technologies in supporting innovation in many sectors: today their application will be shown in non-destructive archaeometric studies and intervention for the preservation and enhancement of Cultural Heritage.

Italy and Hungary are both depository of an extraordinary wealth of Cultural Heritage recognised and envied world-wide. It implies, on the other side, high costs for conservation and preservation; in fact, masterpieces of art and artefacts easily undergo, mainly in our modern times, degradation processes triggered by environmental pollution as well as human negligence and destruction.

Public administrators must be more involved and aware of the new means which are made available by science and advanced technologies. On the other side, experts and scientists gathered here could pave the way to policy makers in undertaking the best strategies of interventions for preserving masterpieces of arts and artefacts of our ancestors.

It will be our duty and the duty of the Italian Embassy in Budapest to help strengthening the bilateral Cupertino which is already on way between Italy and Hungary and promote new intergovernmental projects.

PAOLO GUIDO SPINELLI
H.E. the Ambassador of Italy

The Italian Embassy in Budapest, within its promotional activity for scientific and technological co-operation between Italy and Hungary, has initiated a series of scientific and cultural events illustrating the role which Hungary played in Europe, not least by a celebration of the nation's archaeological and architectural heritage.

On this topic, Italy has a consolidated and long since partnership with Central-Eastern European Countries, mainly with Hungary, for their common cultural roots and unique Cultural Heritage which must be preserved for future generations. Italy and Hungary, in fact, more than other Countries, have the need and duty to preserve this unique Cultural Heritage which belongs to the entire humanity.

The integral connection between science and culture is something strongly felt by the scientists and scholars of both countries; and this has nourished closed and detailed Cupertino, including Cupertino among archaeologists, art historians and architects as it is documented by the many common projects which are already on way between the two countries.

The experts and scientists gathered together in this magnificent ceremonial room of the prestigious Hungarian National Museum will surely pave the way to policy makers in undertaking the best strategies of interventions for preserving masterpieces of arts and artefacts of our ancestors.

In offering my heartfelt thanks to the Hungarian and Italian scholars and researchers who made this major cultural encounter possible, I must also record my satisfaction at the support given by our Embassy in Budapest to an event which clearly illustrates how deeply-rooted are the relations, at this moment in history, between the Italian and Hungarian cultures, not the least in the conservation of the artistic and architectural heritage in Budapest.

On remarking again the high cultural and scientific value of this symposium, I wish you an enjoyable stay and profitable work.

Paolo Guido Spinelli

Ambassador of the Republic of Italy

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ERIKA KONCZ

State Secretary, Ministry of Cultural Heritage

First of all, let me greet all the participants of the Conference in the name of the Ministry of Cultural Heritage, and I would like to render my thanks in particular to the two partner Institutions who took part in the organisation of this event: the Italian Embassy (personally, to H.E. the Ambassador Paolo Guido Spinelli, and to the Scientific Attaché Mr. Benito Righetti), and to the Hungarian National Museum (respectively to the Director, Dr. Tibor Kovács).

Since Hungary's accession to the European Union, the number of international scientific conferences has proliferated, but, as far as I know, on this topic this is going to be the first assembly.

It is a real pleasure for me that Hungary's partner will be exactly Italy in this occasion, because Italy can boost of a great experience in the protection of historic monuments and in the archaeological excavation, elaboration and preservation techniques.

It is well known, that the first archaeological and historic-monument excavations in Italy were initiated by the Princes of the Medici Family, already in the XVIth Century; and during the past centuries Italy has turned out to be a real world-power in the protection of historic-heritage and regarding the scientific methods and technologies that are used in this field.

At the same time, it is less popular that since the XIXth Century also the Hungarian scientists, archaeologists and art historians had joined to this work. Perhaps it is enough to recall to the memories the 'great name' of Torma Károly, excellent researcher of the Roman heritages of Pannonia, university professor and the founder of The Museum of Aquincum, who is also acknowledged in Italy; his tomb is in Anzio, Italy. From the XXth Century, thanks to the foundation of the Hungarian Academy in Rome, the researches had received a severe institutional background and the Academy still carries a very important role in the assistance of our cultural connections.

Regarding the topic of today's Conference, we have to start from the fact that the material part of cultural heritage including national monuments, archaeological sites and artefacts/findings, had accumulated in a large quantity and represent a great value, but they are also finite/restricted, because their formation had happened in the past - which is already closed down- and they can not be produced again as originals.

It is an important feature as well, that the visual information and the sensible context that accompany the 'touchable', '3 dimensional' objects have the same documentary value, and if we 'work out' their meaning they can enrich our scientific knowledge.

However, both the monuments, archaeological sites and artefacts, but even more the accompaniments mentioned after –at least because of their material nature and their age – are extremely vulnerable and temporal/caduceus; their maintenance and preservation for our descendants requires a continuous fight against the laws of nature, physics and chemistry. In addition, many times the research and the scientific examination itself can also expose its subjects to a real danger: although the archaeological excavation can explore indispensable information, the major part of the research processes damages and endangers the monuments.

Because of this, we have to try to invent and use methods, that not only furnish information in bigger quantity and quality, but also reduces the risk of damaging the examined objects and phenomenon. In the same way, we have to find techniques and solutions, by means of which we can fight against the destructive effects of time more successfully and preserve better the discussed monuments. In this battle, the archaeology, the museology, and some natural sciences such as physics, chemistry and the modern technology are allies.

Well, the participants of this Conference are illustrious representatives of this two sides from both countries; their presence is the warranty of common results and process. Let me wish you good luck to this work, and encourage all of you to consider this occasion as the starting point and a station of a process, that will lead to a real Cupertino, that involves institutions and experts.

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LUMINESCENCE DATING AND CULTURAL HERITAGE

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Abstract

Crystalline inclusions present in ceramics act as thermoluminescent dosimeters of the dose due to the natural irradiation field. Because of this various ceramic material (pottery, bricks, cooked clays, clay-cores) can be dated by thermoluminescence (TL). A short review of the main possibilities of TL dating is given, with some examples that underline the advantages and limits of this method in archaeology.

KEYWORDS: THERMOLUMINESCENCE, CERAMIC MATERIALS, ABSOLUTE DATING TECHNIQUES

KULCSSZAVAK: TERMOLUMINESZCENCIA, KERÁMIA, ABSZOLÚT KORMEGHATÁROZÁS

The Thermoluminescence

Thermoluminescence (TL) is the emission of light observed during the heating of insulating or semiconductor materials, provided that they have been previously exposed to ionising radiation (McKeever 1985; Martini e Meinardi, 1997; Chen and McKeever, 1997)

This irradiation may take place in the laboratory or in a radiative environment. Another possibility, which is exploited in dating applications, is when a naturally occurring material is irradiated by the radiation field of its natural surrounding.

The exposure to radiation perturbs the initial stable configuration of the material and the heating allows the release of the accumulated energy.

The existence of thermoluminescence is linked to the internal ordered structure of insulators, and to the presence of imperfections in its lattice. The process can be described, in a simplified way, recurring to the energy band representation of insulators and assuming the presence of two kinds of imperfection in the crystal, as shown in **Fig. 1**.

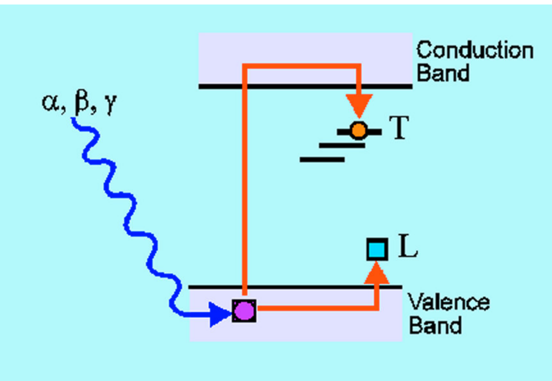

Figure 1. Traps levels in an insulating crystal

As a consequence of the exposure to ionising radiation, electrons and holes (vacancy of an electron) are produced in pairs: they can be captured in the electron and holes traps, whose energies are within the forbidden gap of the crystal. These traps are metastable, and usually their lifetime is very long at room temperature. The higher the exposure to ionising radiation, the higher the number of electrons and holes trapped. In general, linearity is observed between the two quantities.

When the temperature of the crystal is increased, the carriers are raised energetically and freed from their traps to the conduction band from which they can recombine with a trapped electron or hole, thus emitting TL.

The curve representing the intensity of emitted light as a function of temperature is called glow-curve, and its shape and intensity depend on the material and on the characteristics of the irradiation field, mainly the type and energy of radiation and its total amount (**Fig. 2**).

The study of the TL properties of a crystal is actually the study of the imperfections of its lattice,

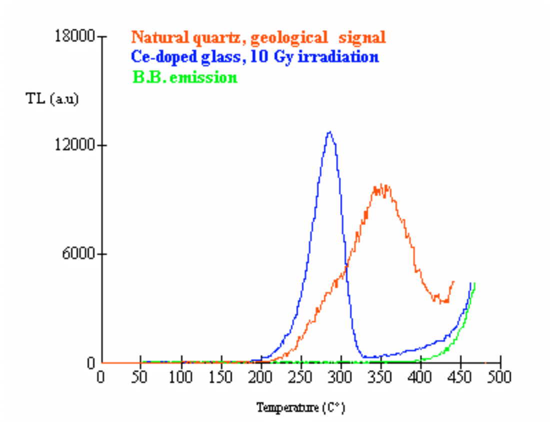

Figure 2. Examples of TL glow-curves

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and TL is a very sensitive tool to detect imperfections even in very small quantities, and to understand the role played by defects and impurities in some physical properties of solids.

This last TL feature mentioned, i.e. its dependence on the amount of the energy absorbed during irradiation, called radiation dose, plays a primary role in the dosimetric applications of TL. (the SI unit for the energy absorbed due to the interaction of ionising radiation with matter is the gray (Gy).corresponding to 1 Joule/kg). In many cases, in fact, the intensity of the TL is directly proportional to the absorbed radiation dose. Once the dose response is tested using calibrated laboratory irradiations, any unknown dose producing a given TL signal can be determined. Several artificial and naturally occurring materials show this favourable property, covering a very wide range of dose (10^{-2}-10^{8} Gy approximately). They are widely used in radiation protection practices and can be used to measure the doses due to occupational exposure and those accrued as a consequence of nuclear accidents. New materials have been developed to best fit the characteristics required by the main specific applications which are personnel, environmental, medical, retrospective and high-dose dosimetry (McKeever et al., 1995).

Detection of TL signal

The definition of TL itself suggests a rather easy way to detect it: what is needed is in fact an apparatus which allows to heat the samples under controlled conditions, and an efficient light detection system. In most cases, the very low level of the emitted signal and the difficulties of controlling and measuring precisely the sample temperature require the use of complex and specifically designed systems. This is particularly true when TL intensity is very low, like in dating applications or when basic studies on defect centres are carried out.

In **Fig. 3** a schematic diagram of a TL measuring instrument is represented. Three main parts can be envisioned: the heating system, the detection system and the signal processing. The most common heating system is composed of a resistive planchet that heats up as a result of the passage of current through it. A common method of measuring the temperature is through the use of a thermocouple welded to the underside of the planchet. A photomultiplier tube (PMT) is normally used to detect the emitted TL.

In fact the efficiency of high-gain, very sensitive PMTs allow the detection of very low level signals with a convenient signal-to-noise ratio.

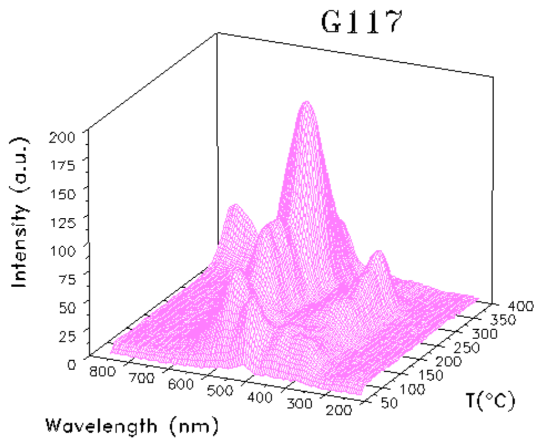

Figure 3.

Ancient mosaic glass. Wavelength resolved TL spectrum

The recent development of high sensitivity light detectors, such as intensified diode arrays and Charge Coupled Devices (CCD), has allowed the measurement not only of the amount of emitted light, but also of its wavelength (Martini et al., 1996), obtaining information about the centres involved in the recombination processes. An example of such spectrum is reported in **Fig. 4.**

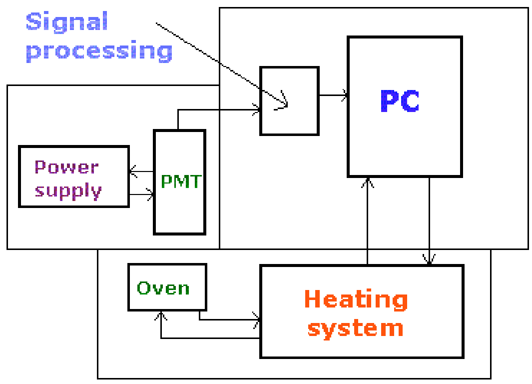

Figure 4.

Diagram of a typical TL measuring system

Thermoluminescence dating

Thermoluminescence dating is the only physical technique for determining the age of pottery presently available. It is an absolute dating method, and does not depend on comparison with similar objects. The application of thermoluminescence in archaeological and geological dating (Aitken 1985, 1990) is based on dosimetry: it stands on the fact that many naturally occurring TL mineral constituents of ceramics, including quartz and most feldspars, are able to act as dosimeters for the amount of ionising radiation they are exposed to. This radiation mainly comes from the radioactive decay of uranium, thorium and potassium present in the ceramics itself and in its surrounding (typically the burial soil), at concentrations of a few parts per million. The radioactive materials having long half lives of 10^9 years or more, the radiation flux is practically constant.

An important point to single out is that, when pottery is fired, it loses all its previously acquired TL. Thus, after cooling, the natural radioactivity causes thermoluminescence to build up again so the older an object is the more light is produced (**Fig. 5**). The TL level measured in pottery is associated with the dose accumulated since it was fired in kiln , unless there was a subsequent re-heating. Any heating at high temperature acts as a *clock resetting* event. This usually occurs when the items are heated over 400°C. In archaeology, thermoluminescence dating is specific for ceramics bricks, cooking hearths, incidentally or deliberately fired rocks such as flints or cherts.

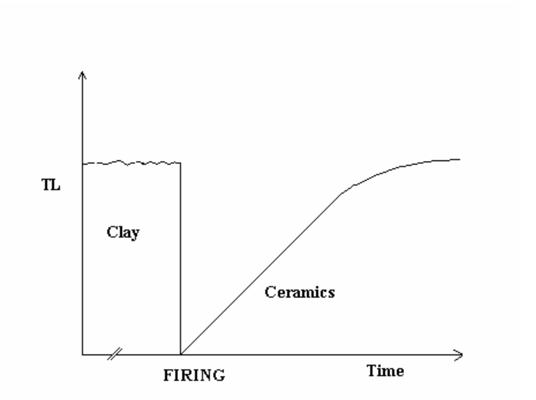

Figure 5. TL growth vs. time

If the radioactivity of the pottery itself, and its surroundings, is measured, the dose rate, or annual increment of absorbed dose, may be computed.

The age of the pottery, in principle, may then be determined by the relation

Age = Absorbed dose / Annual Dose-rate

Typically we are dealing with absorbed doses ranging from a few to a few tens of Gy. The dose-rate is usually within the range 1-10 mGy/year.

Even if the principles on which TL Dating is based are rather simple, the practical procedures are not. The precise evaluation of both absorbed dose and dose-rate requires the consideration of various factors affecting the calculations. For example, one of these is the way in which the different types of radiation, α, β, or γ, are absorbed by the thermoluminescent minerals contained as small crystals in the pottery. The amount of water contained in pottery, changing its density, also influences the absorption of energy.

The main dating techniques were developed on the basis of the differences in absorbing radiation by grains of different sizes. The so called "inclusion" technique (Fleming, 1970) considers only quartz crystal grains in the range 100-200 μm extracted from the ceramic.

The second major TL technique is the "fine grain" (Zimmermann, 1971) which uses all the material that can be extracted, for instance by drilling the sample. A grain size separation is then operated by settling the obtained powder in acetone suspension. It is possible to select a grain size range, typically between 1 and 8 μm.

It must be mentioned that some complicating factors can occur, due to the specificity of the materials. In fact, while in dosimetry one can choose the best dosimeter available for a given radiation, in TL dating only the naturally occurring minerals can be used. The clay minerals have usually low TL; a few of them are hardly thermoluminescent at all; some may not have a straight-line relationship between dose and TL. In addition, some of the accumulated signal may be lost due to thermal and anomalous fading (Wintle, 1973), where part of the TL is lost without thermal excitation, or it may exhibit a spurious, non radiation induced component (Martini et al., 1988). Also, if the sample was poorly fired in antiquity, the TL clock would not have correctly set to zero.

The presence of one or more of these effects has great influence on the uncertainty of the final result. If they are absent or small, or can be compensated or corrected for, then the error limits on the dates obtained typically range from 4 to 8% of the age ($\pm$ 1σ)

Dating applications

TL might in principle be used to date any archaeological material containing thermoluminescent mineral and subjected in the past to an heating sufficient to erase any previous signal. Ceramics, due to its widespread occurrence in archaeological excavations, is the more frequent material submitted, together with bricks from historical buildings.

The clay cores from lost wax metal castings may also be tested. Heated stone material, such as hearths, pot boilers, and burnt flints, can be dated as well, even if some regions are known to present problems for TL, like Indonesia and West Mexico: objects from these areas usually do not successfully yield TL dates, due to the very poor TL characteristics of the raw materials locally used.

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Figure 6.

Bet Gemal excavation site

Possible applications of TL dating beyond man-made artefacts are in geology where aeolian, fluvial, coastal and, in some cases, marine sediments can be dated. In these case the signal resetting is due to the exposure of sunlight during deposition. Also volcanic formations could be dated.

A few examples of application of TL dating techniques in various archaeological fields are reported in detail in the next section.

Excavation archaeology

An interesting example in excavation archaeology is the case of Bet Gemal (Strus, 2003), an Israeli village inhabited from II century BC to the Islamic period (IX century AD).

The site displayed exceptionally well preserved remains: a Roman-Jewish quarter, a Christian Byzantine settlement with several plants for oil and wine production (**Fig. 6**) and an Islamic dwelling place. Each group of remains is related to different chronological periods. The long occupancy of the site and the cultural and religious changes that took place resulted in a complex, cumbersome stratigraphy posing problems of absolute chronology, in particular regarding the duration of the different occupations. TL dating was performed on several domestic ceramics characteristic of the three periods. Supported by our results an absolute chronology of the site could be proposed. The Jewish occupancy had its break at the end of the I century AD, in the historic context of the Jewish-Roman Wars. For the two following centuries, the site should have been almost abandoned until the III century, when the repopulation of the site started and its prosperity grew; the remains of workshops of ceramics, wine and oil presses testify the economical prosperity of this phase.

A successive development of the village occurred in the Byzantine period, linked to new constructions like a church and a further oil press that was functioning during the VI century. The last transformation of the village occurred during the VII century, after a destruction on a large scale probably due to the invasion of the Persian army in 614 AD or to the local Muslim victory over Byzantines in 643 AD. The destruction was followed by a general restoration of life, marked by the re-building of several houses and by new industrial and housing projects, until the final abandonment of the village somewhere in the IX century.

The impressive stone structure depicted in Fig. 6, the bigger of the three oil presses associated to the Byzantine phase, well testifies the economical importance of the site at that time.

Another relevant application is the study of the chronology of the Cham civilisation, that developed in central and southern Vietnam from 6th to 16th century.

In the frame of an Italian-Vietnamise Program of Cooperation an extensive TL dating project of the MySon religious complex started in 2005. The site shows the remains of more than 70 buildings of different styles (**Fig. 7**) built in different periods but always with the same building technique.

About 300 bricks and ceramics have been sampled and the presently available results show evidence of a chronology much more complex than supposed by former scholars, especially for what concerns the important edification phases occurred during X and XI centuries.

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Figure 7.

A ruined tower at the MySon religious complex

Historical buildings

Since the stratigraphic techniques initially developed for archaeology have been extended to architecture, the relative internal sequence of the various building phases of a monument can be usually precisely determined. Their absolute chronology is however sometimes problematic or controversial. In such cases, the contribution of the TL dating techniques could be conclusive (Galli et al., 2002).

It must be reminded, however, that care has to be taken when associating the TL age of a brick to that of the structure it belongs to, because the event that is determined is the last firing of the sample. Voluntary human actions (rebuilding, transformation, decay and restoration) can modify the position of a brick in the stratigraphic sequence of a building. Moreover, in case of reuse of materials from pre-existing structures, dates are older than the building; in case of upkeep or mimetic restoration, dates are younger than the building. In case of fire, this event will be dated.

The contribution of the archaeometric techniques to the study of ancient buildings is anyway very important. The main advantages of this kind of application are the availability of large quantities of material, the homogeneity of environmental radioactivity and the lesser extent of humidity fluctuation.

TL dating in architecture should therefore give uncertainty lower than in excavation archaeology, as confirmed by the statistical analysis performed on about 1300 ceramic samples submitted to our Laboratory for dating over the last ten years (Martini et al., 2001). It could be appreciated that errors lower than 6% are much more frequent when dating buildings rather than excavated samples.

As an example, we report the results recently obtained for the San Lorenzo Church in Milano (**Fig. 8**)

Figure 8.

Back view of S Lorenzo Church in Milano

The cathedral of S. Lorenzo in Milano, the more ancient testimony of roman and palaeochristian architecture in Milan, is a complex architectural structure that shows evident traces of several building interventions often lacking of sure chronological attribution. After performing a detailed stratigraphic analysis on both external and internal surfaces to fix the general building sequences, the different phases were dated with thermoluminescence and radiocarbon.

TL was applied only to unbroken bricks and fictile tubes sampled in several wall structures of the complex. Radiocarbon was used on wooden charcoal scrapes contained in the joint of mortars of walls. In absence of scrapes, calcium carbonate clots found in the mortars themselves were employed. In total, more than two hundred samples were analysed.

The very good agreement of all the results relative to the original phase allowed to indicate the narrow period 390-410 A.D. for the foundation of the tetraconc. It seems to be a more reliable estimation than the previous one based on historical ground. Later medieval reconstruction phases have also been uncovered.

Burnt flints

The possibility of dating burnt flints by TL appeared soon a great challenge to contribute in studying sites whose age is beyond the upper limit of radiocarbon dating (about 40.000 years), and when organic materials are not abundant or not well preserved. Flint is dense siliceous sedimentary rock whose basic component is SiO_2, occurring as silica, cristobalite/trydimite and α-quartz.

Figure 9. Archaeological flints

Due to its hardness and conchoidal fracturing properties, it was largely employed in prehistory to manufacture a large number of artefacts (**Fig. 9**). Some of them were accidentally or deliberately heated and the burning is obviously essential for the erasure of the geologically accrued TL.

Goksu and co-workers (Goksu et al., 1974) highlighted the possibility of dating burnt flint, presenting at the same time the limits and the specific problems related to such materials: generally low TL sensitivity and sensitivity changes, spurious and regenerated TL and very low concentrations of radioactive elements, circumstance that attaches great importance to a precise evaluation of the ambiental dose-rate. Despite the problems encountered in this application, flint dating is widely used and the results played, for instance, a primary role in the revision of the chronology of the presence of Neanderthal man and of modern human in the Middle-East.

We recently studied a group of 20 burnt flints from the prehistoric site of Fumane, in North Italy, Verona province (Martini et al., 2001). It is a huge cave, used as a shelter by ancient men, characterised by paleosurfaces extremely rich in bones and lithic objects. The study of this site is considered very important for the passage from Middle to Upper Palaeolithic and from Mousterian to Aurignacian age in Northern Italy and Europe. Some stratigraphic sequences of the site have been dated with radiocarbon but very few data regarding the human presence are available.

The chronology obtained by TL, spanning from 79 $\pm$ 11 ka to 57 $\pm$ 12 ka BP, added key information to the archaeological and palaeoenvironmental history of this Pleistocene period, up to now poorly dated.

Archaeological glasses

The chemical-physical behaviour of silica glasses suggested the use of TL techniques as a suitable method to date these materials. Actually, because of the amorphous nature of glass, numerous factors reduce its thermoluminescence sensitivity. The main problems encountered in glass dating are generally low TL sensitivity (TL emission per unit of dose) and the emptying of TL traps due to sunlight exposure or to the low stability of TL traps at room temperature. Both effects result in a loss of TL signal. Moreover, changes in TL sensitivity often occur after repeated heating and irradiation of the same sample. Due to these difficulties, at the present state of the art only a few percent of the samples analysed could be dated.

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Figure 10.

Byzantine mosaic glass *tesserae*

We focused our attention to a particular class of glass, the vitreous *tesserae* composing mosaics (**Fig. 10**). Our study was performed on samples chosen as representative of six sets of differently coloured glass mosaic tesserae. They all belong to wall mosaic decorations and were found in archaeological excavations or taken from mosaics to be restored, all well dated on archaeological grounds.

The thermoluminescent emission of these vitreous materials, lacking a long range periodic structure, is due to the impurities present or added to the glass network (Al, Mn, Cr…), the colour centres acting as electron traps and recombination centres. In fact, a good natural TL emission was observed in almost all tesserae, the blue ones being generally characterised by higher sensitivity.

Samples were submitted to different protocols for TL measurements, previously described (Chiavari et al., 2001) and their TL properties were investigated. This allowed selecting eight tesserae characterised by suitable TL behaviour (high sensitivity, trap stability, low optical bleaching and limited changes in sensitivity after heating), that were submitted to dating. They presented a TL sensitivity comparable with that of ceramics materials.

For the external annual dose-rates, mean values typical of the different provenance areas have been assumed, with errors taking into account possible wide variations. These assumptions determined errors quite high (15-18% of the age). It is however noticeable that the TL dates are generally consistent with the archaeological ones. It is also remarkable that we could date eight tesserae over the nineteen analysed: the percentage of suitable samples was about 40 % against the 5% reported for glasses up to now (Chiavari et al., 2000).

Clay-cores

The first application of TL techniques to clay-cores dates back to 1974, when D.W. Zimmermann (Zimmermann et al., 1974) succeeded in testing the authenticity of core materials from a Bronze Horse of the New York Metropolitan Museum of Art. Further attempts devoted to dating, soon enlightened a series of difficulties, complications and limiting factors.

First of all, the application is in principle possible only for the objects cast by lost-wax technique, using the remains of thermoluminescent clay-cores heated contemporarily to the casting itself. The possibility of dating such materials depends on its mineralogical composition, and particularly on the abundance of “good” thermoluminescent minerals like quartz and feldspars. A high concentration of carbonates and/or organic material is generally a disadvantage, for the associated spurious, not dose-dependent TL emission. Another phenomenon that is observed with higher frequency than in ceramics is the anomalous fading, a process which empties deep traps at room temperature. The evaluation of the environmental contribution to the annual dose-rate can be problematic, both for the often unknown “archaeological history” of the object to date and for the need to evaluate the shielding effect of the bronze layer on the external irradiation.

Due to the sum of these circumstances, the achievable precision in dating bronzes is generally lower than in ceramics, the mean error being generally about $\pm$10% of the age.

As a further remark, it must be pointed out that dating the clay core is not dating the bronze statue itself, except when the correlation between the ages of the two objects is sure, or highly probable. It must be reminded that any TL dating refers to the last heating at high temperature experienced by the item to date: in case of restoration or repair performed by heating, this last event will be dated instead of the original one (Martini and Sibilia, 2003).

The possibility of dating clay-cores is furthermore precluded if the object has been intensively radiographed before sampling out the core material. In such a case, unfortunately frequently recurring, the high energy X-ray exposure results in an accrued radiation dose that produces an additional TL emission, superimposed on the archaeological one. The evaluated palaeodose is consequently meaningless.

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Figure 11.

Benvenuto Cellini, Perseo

Nevertheless, things are not always so discouraging, and often very satisfactory results can be obtained, like in the case of the Cellini's Perseo (**Fig. 11**) Benvenuto Cellini (Florence, 1500-1571) wrote that a "great cry of admiration" arose from the throng gathered to watch the unavailing of his *Perseo* in the Loggia dei Lanzi on April 27th, 1554. After about five hundred years of open air exposure, the state of conservation of the statue was critical, due to the polluted, aggressive urban atmosphere, transforming its historical patina from insoluble to soluble salts. It was therefore fully restored and its disassembling gave access to its interior, where important fragments of clay-core were found. In this case, TL dating was mainly performed to check the reliability of the technique, being the dating itself beyond dispute.

The dating result, 1540±35 AD, is in very good agreement with the historical records, confirming the potential of such application, the reliability of the laboratory protocols and the accuracy and precision of instrumental calibrations.

Conclusions

TL dating of ceramic materials is nowadays a consolidated and powerful technique which supports the archaeological and archaeometric researches.

Precision as good as ±5% in the evaluation of the age of various kinds of archaeological findings are often reached, allowing the solution of archaeological or historical problems arising from samples chronologically relatively close.

A systematic comparison of TL dating results with those obtained by other absolute dating techniques like radiocarbon and dendrochronology and the dating of samples already well independently dated on archaeological or historical grounds is highly recommended, in order to check and improve precision and accuracy of the laboratory experimental procedures.

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NON-DESTRUCTIVE INVESTIGATIONS OF CULTURAL HERITAGE OBJECTS WITH GUIDED NEUTRONS: THE ANCIENT CHARM COLLABORATION

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Abstract

When investigating valuable artistic objects, the first and foremost requirement is to preserve the integrity of the objects. Various kinds of physical processes can provide information about the material of the objects, without destroying them. Neutrons, elemental particles having zero electric charge can enter deep into the irradiated material, and they can undergo different nuclear interactions. Both the neutron capture-based methods (i. e. Neutron Tomography – NT, Neutron Activation Analysis – NAA, Prompt Gamma Activation Analysis – PGAA and Neutron Resonance Capture Analysis – NRCA) and the neutron scattering-based methods (i. e. Time of Flight Neutron Diffraction – TOF-ND and Small Angle Neutron Scattering – SANS) have been previously applied to archaeometric research. A new European Commission funded project with ten collaborators, called Ancient Charm, has been launched with the aim of combining the aforementioned methods to achieve 3-D imaging and elemental mapping of museum objects with complex structures.

KEYWORDS: PROMPT GAMMA ACTIVATION ANALYSIS, NEUTRON RESONANCE CAPTURE ANALYSIS, TIME OF FLIGHT NEUTRON DIFFRACTION, 3-D IMAGING, ANCIENT CHARM

KULCSSZAVAK: PROMPT GAMMA AKTIVÁCIÓS ANALÍZIS, NEUTRON REZONANCIA BEFOGÁSOS ANALÍZIS, NEUTRON DIFFRAKCIÓS VIZSGÁLAT, HÁROM DIMENZIÓS KÉPALKOTÁS

Introduction

Archaeologists search for material clues of ancient cultures, trying to answer the most frequently emerging questions: When was a cultural object produced? Where, in which workshop was it produced? Was it made of local or imported raw material? Is it really a significant one? Classical archaeologists perform their research mainly on the basis of the stylistic features of the artefacts. On the other hand, already since the dawn of the modern science, geologists, physicists, chemists and biologists tried to apply the experiences of their own disciplines on Cultural Heritage research. When investigating a valuable artistic object, the first and foremost requirement is to preserve the integrity of the object. Archaeologists and curators usually don't allow any damage to be done to the artefact, even if it is a non-visible, micro-destructive one.

Fortunately, various kinds of physical processes can provide information about the materials of the objects, without destroying them. Neutrons, elemental particles having zero electric charge can enter deep into the irradiated material and they can take part in two basic kinds of nuclear interactions. The first one is the neutron capture, when the neutron is bound into the atomic nucleus. The so called 'capture state' is an exited state of the newly formed atomic nucleus. The probability of the capture (*i.e.* the neutron capture cross-section) depends on the neutron energy, and shows a great variation from one isotope to another. Captures of thermal or subthermal (typically meV energy) neutrons are called thermal neutron capture processes, while higher energy (so called epithermal) neutrons take part in resonance neutron captures. Following the capture reactions, the nuclei emit characteristic γ-photons. Measuring the energies and intensities of the characteristic γ-radiation, the elemental (isotopic) composition of a sample can be determined. These kinds of analyses are called thermal neutron activation analysis (NAA) and neutron resonance capture analysis (NRCA), respectively.

Besides capture reactions, thermal neutrons can be scattered on crystal planes or by individual nuclei. Thus it is possible to obtain a neutron diffraction pattern of a sample, which will reveal information on the crystalline structure of a material, *i.e.* mineral phases, texture or porosity, similarly to X-

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ray diffraction. The neutron scattering process is utilised by neutron diffraction (ND) or by small angle neutron scattering (SANS).

When detecting the transmission of the impinging neutron beam we can record a picture of the object with clearly distinct parts of different neutron absorptions. This is the basis of neutron tomography (NT), similar to X-ray tomography. In parallel with the development of large experimental facilities (research reactors and accelerators), all the above mentioned neutron-based methods have already been applied in archaeometric research. Instrumental Neutron Activation Analysis can be regarded as routine method, while applications of Neutron Resonance Capture Analysis and Neutron Diffraction in archaeometry are quite new (Blaauw et al. 2005 and Botti et al. 2006). However, combination of them in order to extract as much archaeologically relevant information as possible is a completely new idea. This is to be worked out in the frame of a European Community founded project, called *ANCIENT CHARM*.

Discussion

What is ANCIENT CHARM?

The title of the project: ANCIENT CHARM – is an acronym for **A**nalysis by **N**eutron resonant **C**apture **I**maging and other **E**merging **N**eutron **T**echniques: new **C**ultural **H**eritage and **A**rchaeological **R**esearch **M**ethods. The project started in January 2006 and will last for three years. Its total budget is about 2 million Euros. Ten European institutions participate in it, namely the *Universitá degli Studi di Milano-Bicocca*, the *Universitá degli Studi di Roma Tor Vergata*, the *Reinische Friedrich-Wilhelms Universität Bonn*, the *Universität zu Köln*, the *Technical University Delft*, the *Leiden University*, the *Institute for Reference Materials and Measurements* in Geel and the *Central Laboratory of the Research Councils* in Didcot. Two institutes are involved from Hungary – the *Hungarian National Museum* and the *Institute of Isotopes of the Hungarian Academy of Sciences*.

The aim of the project is 'To provide a new, comprehensive neutron-based imaging approach, which will be applied for the 3-D imaging of elemental and phase composition of objects selected as a result of a broad scope archaeological research'. The tasks have been organised into workpackages, which will be performed by various working groups.

The research institutes, which exploit the benefits of large facilities, offer their technical support to develop new methods for Cultural Heritage research. On the other hand, as a starting point, museum experts will help to construct laboratory model objects, which are similar to the actual real-life archaeological ones from the point of view of the applied methods. Later they will select a representative collection of scientifically interesting objects which are worth to be investigated.

The experiments will be conducted in the following large facilities: *1.* The Prompt Gamma Activation Analysis (PGAA) facility at the Budapest Research Reactor, which operates with cold neutron source since 2000. The current neutron beam intensity is $5\cdot10^7$ n/cm^{2}s; *2.* The PGAA and ND facility at the FRM-II Research Reactor in Garching, Germany. A neutron beam of 10^9 n/cm^{2}s will be available from 2007. *3.* The pulsed neutron beam of GELINA, Geel, Belgium, is gained from a 150 MeV LINAC accelerator, which has already been used for NRCA. *4.* The 800 MeV pulsed spallation neutron source at ISIS, UK, is regularly used for Time of Flight Neutron Diffraction (TOF-ND) experiments on archaeological objects.

From PGAA and PGAI

We apply Prompt Gamma Activation Analysis in archaeometry research at the Institute of Isotopes, Budapest since 1997. At an early stage, we had a thermal beam of $2.5\cdot10^6$ n/cm^{2}s. Following the set-up of the Cold Neutron Source in 2000, we utilise a cold neutron beam of $5\cdot10^7$ n/cm^{2}s thermal equivalent flux. We have successfully applied the method for a provenance study of prehistoric chipped stone tools made of obsidian (Kasztovszky & Biró 2004), flint and porphyry (Markó et al. 2003), a provenance study of Neolithic polished stone tools made of basalt, greenschist and blueschist (Szakmány & Kasztovszky 2004), and we have searched for the raw materials of Venezuelan pre-Hispanic pottery (Kasztovszky et al. 2004). We characterised baroque glass objects (Kasztovszky et al. 2005), and also Roman and Greek bronze objects (Vaday et al. 2002), as well as Roman silver coins (Kasztovszky et al. 2005). We have built good scientific co-operations with the Hungarian National Museum, and with foreign Institutions (Tübingen University, Simon Bolívar University of Caracas, Institute of Nuclear Chemistry and Technology in Warsaw, to name a few).

The elemental identification with PGAA is based on our standardisation method (Révay & Molnár 2003). The instrumentation and the concentration calculation are also published by Szakmány & Kasztovszky 2004. In archaeometry research the absolutely non-destructive feature is highly capitalised. After a few days of decay there is practically no residual radioactivity, and there is no damage to the object.

PGAA, which applies a 20-400 mm^2 cross-section beam, is definitely a bulk analysis method. Usually, the detected major- and trace components – as average values – provide reliable information to characterise homogeneous objects, such as stones, glass and metals. On the other hand, archaeologists possess numerous complex and composite objects with many fine details inside or on their surfaces, which should be informative to analyse. How can we 'focus our eyes' on such small components, how can we map the compositions with our neutron beam?

With a few millimetre-sized neutron beam, the emitted γ-photons are also detected from a limited part of the object. This is how we can 'cut' and analyse a certain part of the body of the object. Moving the object in front of the beam – practically rotating around a vertical axis and translating vertically and horizontally with an automated system – we can map each part of the object. PGAA measurement of each section will result in one individual spectrum, but one has to keep in mind that reducing the beam size will result in loosing the intensity, thus the data acquisition time might be considerably longer. From the step of collecting the required spectra, it is 'only' computer work – although not an easy one – to construct the three dimensional elemental 'map' of the object. This technique is called Prompt Gamma Activation Imaging (PGAI). To perform complete 3-D mapping of a whole object with PGAI, however is very time consuming, thus it is plausible to combine it with the much simpler neutron tomography (NT), which, in turn can provide a resolution of some 10 µm. The idea is to get an overall picture of the object with NT first, and then to focus on the sections of interest with PGAI.

In 2005 we have performed a test experiment on a simple artificial object. A cylinder of 10 mm diameter was filled with SiO_2 (equivalent to sand), and a 1 mm thick copper rod was placed next to the inner wall of the cylinder. The sample container was made of Teflon, producing very low background signal, which was taken into correction. The sample was rotated around its vertical axis, in front of a neutron beam, collimated with 1 mm wide vertical slot (***Fig. 1a***). From the measured PGAA spectra we checked the intensity of significant Cu-peaks (***Fig. 1b***).

The peak intensities have varied significantly with the angle of rotation, which indicates that the 3-D elemental mapping is possible, not only in theory but also in the practice. Furthermore, the above described principle is much more complicated in the reality. As we mentioned, the atomic nuclei not only capture but also scatter the neutrons. The ratio between the capture and scattering events depends on the ratio of the absorption and the scattering cross-sections. In such materials where the scattering effect is large (typically H- and C-containing, *i.e.* organic ones) the pencil-beam will be widen, thus the image of a particular part will be blurred, as for a photo made in fog. On the other hand, details behind a highly absorbing material (one practical example is silver) can not be detected, because the nuclei on the front will capture most of the neutrons within a very short distance. In case of silver this critical distance is around 1 cm.

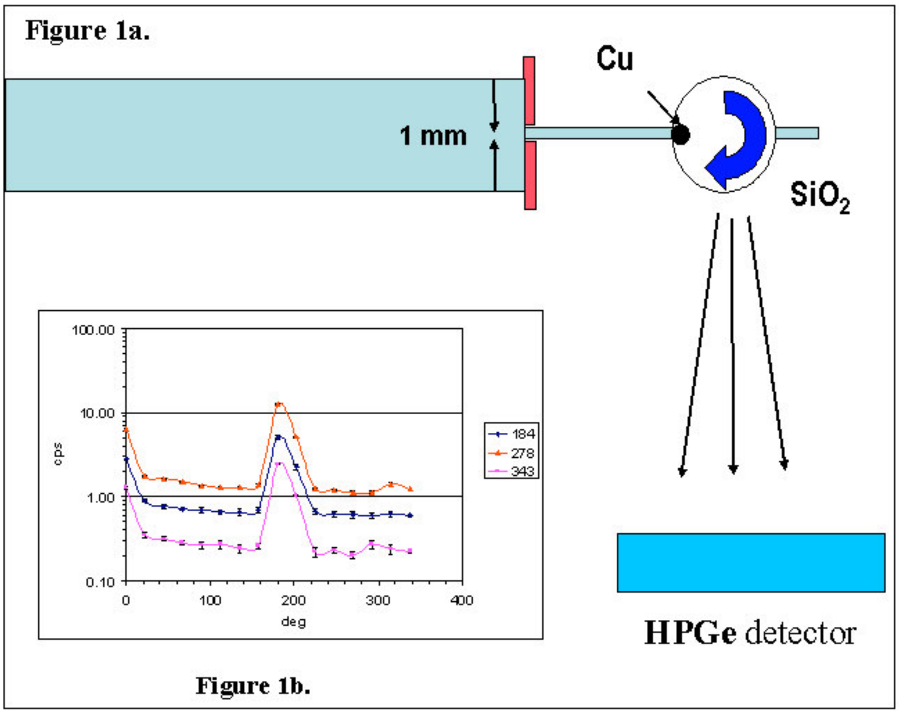

Figure 1a.

Schematic layout of a pilot measurement for Prompt Gamma Activation Imaging (PGAI) – performed at the Institute of Isotopes, Budapest

Figure 1b

Intensities of 3 characteristic prompt-γ lines of copper (185 keV, 278 keV and 343 keV) as function of copper rod position. The intensities are given in counts per second, while the positions are given in degrees.

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It can be seen that prior to the measurements of real objects, one must perform simulation calculations to choose the feasible compositions from the everyday practice, and also one need to run test measurements on simple but relevant reference model objects made in the laboratory. Already in this phase of work the close co-operation between scientists and museum experts is a strong demand.

The next step: Analysis of actual Cultural Heritage objects

Following (or parallel with) the basic methodological considerations, we have to select the most important, most characteristic set of objects in order to obtain the highest level information with reasonable efforts. The principles of selection should fulfil some basic criteria: *1.* Current archaeological research problems should drive the selection of objects, which, in turn, set the measurement requirements. *2.* The selected objects should be representative of different classes. Possible choices are objects with voids; composite materials; composite objects containing a core; jewellery with inlays and multi-layered objects, *etc.* *3.* Other selection criteria: They should be representative from the point of geographical provenance (throughout Europe) and from the point of archaeological, historical periods (*e.g.* Neolithic, Roman, Early medieval, *etc.*). *4.* Finally, the information output from neutron-based methods should be unique.

An initial collection has been selected (by Zs. Hajnal and K. Biró) comprising the following archaeological objects from the Hungarian National Museum:

1, DISC FIBULA WITH ALMANDINE INLAYS (Kölked, Hungary; 2nd half of 6th c. AD – ***Figs. 2a and 2b***)

Disc fibulas with almandine inlays had been imported into the territory of the Avar Empire. They originated from the Frankish settlement area. Among these types, the main iron structure with silver or gilded silver covering plates is very rare (Kiss 1996), they supposed to be the production of Merovingian workshops. With the help of 3-D elemental mapping (PGAI) of the fibula we would like to answer the following questions:

1, Whether the main structure was made of gilded silver and, consequently whether the iron band could be a repair part, or whether the object belongs to the rarer group of iron disc fibulas.

In the first case, the object could be a local product, which was later repaired in the Avar period. In the latter case, it is very likely that the fibula is a western import from North-West Germany or from North-East France.

Figure 2a and 2b. Photo of the disc fibula with almandine inlays, from the collection of the Hungarian National Museum (origin: Kölked, Hungary; 2nd half of 6th c. AD, Grave A 279; 76.1.45). This object became the logo of the Ancient Charm project.

Fig 2a: front view, Fig 2b: lateral view.

2, What is the material of the filling material of the cells below the almandine plaques? *3*, It would be interesting to know the exact form and the material of the white pearl in the middle.

2, IRON BELT MOUNTS (Környe, Hungary, 1st half of 7th c. AD – ***Fig. 3***)

The objects are representatives of a three parted Merovingian type collection (dreiteilige Gürtelgarnitur) (Salamon & Erdélyi 1971).

Figure 3. Photo of the iron belt mounts, from the collection of the Hungarian National Museum (origin: Környe, Hungary 1st half of 7th c. AD, Grave 88; 69.1.205)

They are triangular and rectangular iron belt mounts with silver messing and glass inlays decorated with geometrical motifs. According their decorations the belt mounts are definitely local products. It is interesting that glass inlays occurred rarely in this type of iron belts. The glass inlays have deep engraved cells surrounded by copper or brass bands. They might be filled with special cement-kit and could contain gold foils under the glass plaques. The way of the silvering and brassing, as well as the depths and the correct materials of the inlays should be investigated. By identifying the similarities between the Western and the local production techniques, we may draw conclusions regarding the origin of the Merovingian type culture.

3, IRON BELT MOUNTS AND STRAP ENDS (Kölked, Hungary; last third of 7th c. AD – ***Fig. 4***)

These objects are iron belt mounts and strap ends from a local-made, but Merovingian type multiple belt garniture (vielteilige Gürtelgarnitur) with S-formed decoration (Kiss 1996). The material might be partly bronze: the special soldering material could contain large amount of bronze. According to X-ray radiography, some other type of filling material could be observed among the iron or bronze parts. This filling material supposed to be wax, glass or enamel, what could be decided by PGAI.

Figure 4.

Photo of the iron belt mounts and strap ends from the collection of the Hungarian National Museum (origin: Kölked, Hungary; last third of 7th c. AD, Graves B 80 and A 454; 78.2.20, 78.2.24., 87.1.38., 87.1.40.)

Final remarks

The Ancient Charm project is one step towards combination of the existing neutron-based techniques in order to apply them in archaeometry research. Extension of the bulk analysis towards 3-D elemental mapping and imaging will raise the amount of valuable information about museum objects with finer details. The project, however, is only in the starting phase yet; some basic issues have to be decided first. Based on model calculations and pilot measurements on artificially made test objects, we have to choose the most appropriate actual objects of interest. All steps, the selection part, the construction part, the data collection, as well as the interpretation of the results require strong and permanent collaboration between museum experts and scientists.

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ION BEAM ANALYSIS AND RADIOCARBON DATING

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Abstract

The working principles and main characteristics of the two main categories of nuclear techniques for applications to Cultural Heritage problems, i.e. Ion Beam Analysis for compositional characterisation of materials and Accelerator Mass Spectrometry for ^{14}C dating of archaeological finds, are briefly recalled. The new Florence Tandem accelerator laboratory is presented and some applications described, as examples of the great potential of these techniques in the specific field.

KEYWORDS: NUCLEAR ACCELERATOR-BASED TECHNIQUES, ELEMENTAL ANALYSIS, ARCHAEOLOGICAL DATING

KULCSSZAVAK: NUKLEÁRIS GYORSÍTÓ ALAPÚ TECHNIKÁK, ELEMÖSSZETÉTEL VIZSGÁLAT, RÉGÉSZETI KORMEGHATÁROZÁS

Introduction

The most important contributions of "nuclear" techniques for archaeometrical studies are undoubtedly the analysis of materials (by X Ray Fluorescence and Ion Beam Analysis [IBA]) and the dating of finds by ^{14}C (radiocarbon) measurements.

Let us start from the former and first of all let's clear our mind from the idea that material analysis in archaeometry be just a sort of diagnostic obsession of scientists with no importance for the world of humanists. Quite the contrary: it is the answer to specific demands from that community. Historians of art, archaeologists, historians of science and technology can learn a lot from the knowledge of the materials used: both through the information concerning specific works, specific artists or their artistic environment (e.g. for attribution purposes) and to achieve more general knowledge on historical trends, sources of supply of raw materials, existence in the past of trade routes from countries far away, etc.

Equally important, it is the conservators and restorers that need to know the materials of the original art works, both to detect their degradation and to consciously choose techniques and materials to be used in a restoration procedure.

Ion Beam Analysis (IBA)

Ion Beam Analysis is the most sophisticated and complete of material analysis techniques. It is based on the use of beams of particles, obtained by accelerators, to "bombard" the objects to be analysed, used as a target. The interactions of beam particles with atoms and nuclei of the target induce emission of radiation (X rays, gamma rays, primary backscattered or secondary particles) with energies characteristic of the emitting atom or nucleus. It is thus possible to reconstruct the composition of the bombarded material, by collecting and energy-analysing the induced radiation. The great analytical power of IBA derives from the fact that one can easily change beam energy, intensity, size, in order to answer the specific problem (each case is in a sense "special" and may require to adapt the technique to the specific demand). Extended performance is also achieved by simultaneously exploiting the different "signals" (the various kinds of emitted radiation) that provide complementary information. Thus, a very broad, often complete, set of data can be achieved in a single measurement, which is typically very fast (100-200 seconds). Among IBA techniques, PIXE (Particle-Induced X ray Emission) exploits the emission of characteristic X rays from atomic elements, PIGE (Particle-Induced Gamma ray Emission) relies on the emission of characteristic gamma rays from nuclei, RBS (Rutherford Backscattering Spectrometry) detects the energy of beam particles after elastic scattering from nuclei (their energy depends in fact on the mass of the scattering nucleus). The most "popular" process is PIXE, just because it is the most probable of beam-target interactions, deriving from an atomic rather than a nuclear collision (and atoms are a much lager target than nuclei for the particle beams! In more rigorous scientific terms, this is expressed by the much larger X ray emission cross sections with respect to all other interaction cross sections).

In Florence, our group started doing IBA already in the first half of the Eighties, using an old single-ended Van de Graaff accelerator "inherited" from basic nuclear physics activities. We got some successful results of non negligible relevance in different fields like the study of environmental pollution, petrologic research and just the applications to Cultural Heritage.

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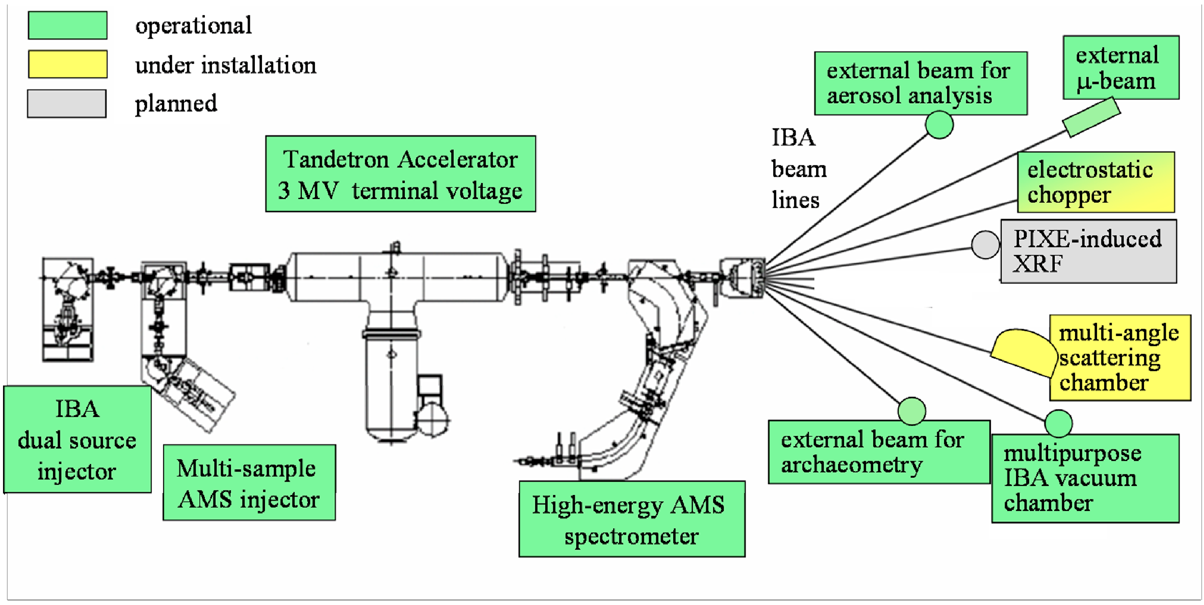

Figure 1. Present layout of the accelerator hall

Also thanks to these results, when recently the Physics Department of the University moved into a new campus where large space and newly built infrastructures were available, the National Institute of Nuclear Physics (INFN) decided to fund the installation of a dedicated laboratory, mainly for applications to C.H. The heart of the new laboratory, named LABEC (LAboratorio di tecniche nucleari per i BEni Culturali) is a new 3 MV Tandetron accelerator, specifically designed to perform both IBA and Accelerator Mass Spectroscopy (AMS), in particular radiocarbon dating. The project started at the end of 2001 (while the building, funded instead by the University of Florence, was still in the early stage of construction); thanks to a hard work of our group and of many more people both in the University and in the INFN management, the installation of the accelerator started already in May 2003, and after one year we were operational with both IBA and AMS. The present layout of LABEC (December 2005) is shown in **Figure 1**, with AMS fully operational, four IBA beamlines already active and a fifth almost completed, plus two more planned to be installed within the next couple of years. **Figure 2** shows a view of the accelerator hall.

It is essential, for a laboratory willing to perform IBA applied to C.H., to be provided with an external beam set-up. We have been using external beams for IBA since the very first activities in the Eighties, developing more and more complete set-ups including more detectors for PIXE and other techniques. The reason why an external beam is a must for C.H. applications is that by such a facility you can investigate in a non-destructive way the complete quantitative composition of any material you may be interested in, with great ease in handling the "targets" (often fragile, sometimes also rather large!). During over twenty years of experience, we have been analysing hundreds of samples or whole works of glass, ceramics, ancient illuminated manuscripts, historical documents, drawings, paintings on wood or canvas. References to these works, as well as to all the past and present activities of LABEC, can be found in the website of the laboratory, http://labec.fi.infn.it[1]

Let me here briefly mention just the analysis of tens of hand-written notes by Galileo concerning the problems of "natural motion", which are a sort of record of his way of thinking throughout his life, from wrong beliefs to the final modern achievements that he reported in the printed works. It is of the utmost interest to reconstruct and locate in time the steps of his "conceptual path" towards these achievements, but these notes are not dated and it is not obvious to derive their chronology from just their content; the idea was then to provide a support to the historians' hypotheses through the analysis of inks. They were not a standard industrial product at Galileo's times, so that each time a batch of ink was over, the new ink (self-prepared or bought from shops) although being made of the same ingredients was never quantitatively equal to the previous. In other terms, we showed that ink composition can be correlated to the period when it was used, and by the matching of ink composition in dated documents (we analysed a large number of dated writings, e.g. letters) and in the undated notes, a chronological reconstruction of the latter was in several cases possible.

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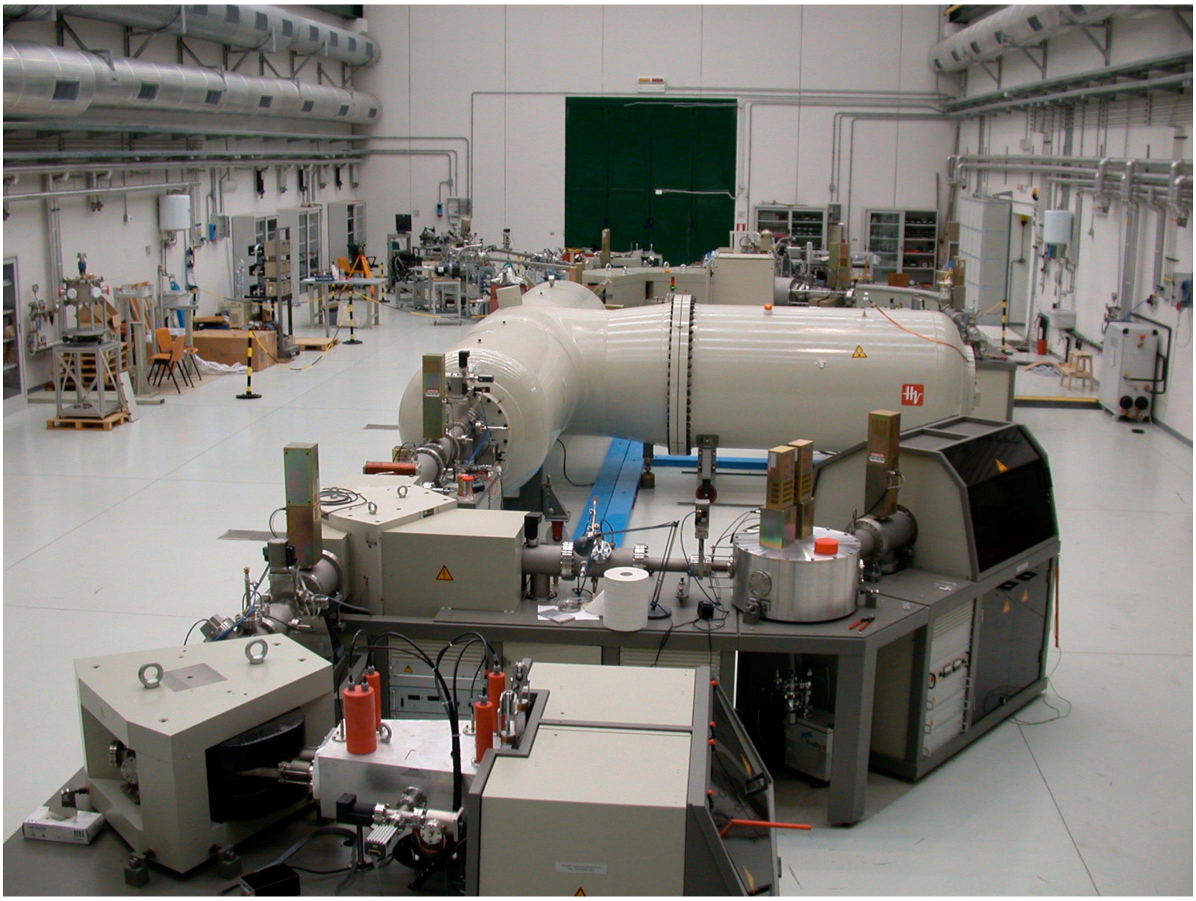

Figure 2. Overall view of the LABEC accelerator hall

Among our IBA measurements for C.H., another case that I wish to mention is the analysis of a famous painting by Leonardo, the Madonna dei fusi (ex-Reford version). In the analysis of materials in paintings on wood or canvas, in general, the analytical problem is connected with the layer structure of these works, and with the presence of the surface protective varnish, if it cannot be removed (as in the case of this painting; sometimes instead the varnish can be removed, when e.g. a restoration must be performed). The problems are:

1) within the beam-induced radiation, you cannot discriminate which come from the varnish, or from the paint layers (you may also have a superposition of more), or from the *imprimitura* and painting preparation. All signals are mixed in the radiation energy spectrum; in particular, one is usually NOT interested in varnish composition, while it is the paint layers that one would like to characterise. Thus, the varnish contributes a sort of undesired background;

2) the surface layer of varnish absorbs the X rays of lower energy (associated to low-Z elements) produced by the beam penetrating into the paint below. Thus, low-Z elements in the paint layers remain undetected by PIXE, while for the identification of some pigments (e.g. lapis-lazuli) it would be essential to detect them: for instance, in the case of lapis-lazuli, the detection of sodium would be a sort of fingerprint of its presence.

As to point 1), the solution has been found by applying what may be called "differential PIXE", which consists in performing measurements on the same area with beams of different energies. At different energies, beam ranges are different and probed depth also changes, thus by comparing X ray spectra taken at different energies, stratigraphic information can be obtained.

As to point 2), the solution comes from using PIGE simultaneously with PIXE. Gamma rays, even though produced below the varnish, are not absorbed since their energy is much higher than the one of X rays. Since it is just from the low-Z elements that gamma ray emission can be more easily induced, the combined use of PIGE and PIXE is of great use to guarantee the detection of low-Z elements even when they are "buried" below some surface layer.

Both ideas were applied in the analysis of the Madonna dei Fusi, leading to a complete characterisation of Leonardo's palette in this work and to the reconstruction of the layer sequence: it came out e.g. that Leonardo's technique was so

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delicate that ultra thin layers of only 10 to 20 micron thickness were laid to produce chromatic shades!

The most recent implementation of our laboratory in the way to perform IBA has been the use of beam scanning procedures on the work to be analysed, in order to reconstruct the concentration map of the various elements. Indeed, most of the materials in the field of C.H. are inhomogeneous, and details of small size or inhomogeneous structures in the order of 100 µm or so are not always easily recognised by visual inspection. From single-spot measurements, the risk of misleading information exists: using too broad beams, one may mix information referring to different materials, using too small beams, one might inadvertently analyse anomalous, non-representative "points". For this reason, in the standard way of performing IBA, measurements are always repeated on several points assumed to be of equal composition, to check how homogeneous is the structure.

A really dramatic improvement in significance, reliability and completeness of information can now be obtained, using methodologies providing "compositional maps". This can be e.g. realised by the scan of relatively large areas (~ some mm^2) with beams around 100-200 µm size, using a data acquisition system that not only records energy signals from detectors, but associates them to the instantaneous x-y position of the beam on the target when the signal was produced. The element-characteristic energies of X rays, gamma rays, etc. are thus coupled to the "pixel" from which they originated on the scanned area, and stored as energy-x-y triplets in the computer memory (list-mode acquisition). It is subsequently possible to replay the acquired data and reconstruct concentration maps of the detected elements. Then, by selecting *a posteriori* homogeneous sub-areas within the scan in such a way as to avoid the compositional "anomalies" discovered thanks to the maps, one reconstructs "safe" energy spectra from which the composition of really representative areas is quantified.

Scanning IBA should always be preferred to single-point measurements, whenever possible. Sometimes it is really essential; an example is in the analysis of metal-point drawings on prepared paper, a drawing technique extensively used by the greatest Renaissance painters in Italy and Europe.

Knowledge of materials (the metal alloy used for the drawing stylus and the pigments used for the preparation of the coloured substrate paper) is needed for conservation purposes: one is dealing with very fragile and precious works, so far little studied, and mainly from the art-historical point of view.

Figure 3. Track left by a Pb stylus on paper prepared using a mixture of cinnabar (HgS) and lead-white (($2PbCO_3$ $Pb(OH)_2$)

There is a problem however: the track left by the metal stylus is not uniform as can be seen by **Figure 3**, and - especially when the paper is prepared using compounds of the same metal used for the stylus - it is difficult to discriminate the track from the paper and quantify the composition of both. By compositional mapping (through the scan over an area of the drawing) the problem is solved and any ambiguity is eliminated (**Figure 4**).

Radiocarbon Dating

Let's briefly talk also about radiocarbon dating. The principle is very well known: the age of an archaeological find (of organic origin, such as wood, seeds, burnt carbon, cloth, remains of an animal or of a man, etc.) can be obtained by measuring the residual concentration of isotope 14 of carbon with respect to the stable isotope 12.

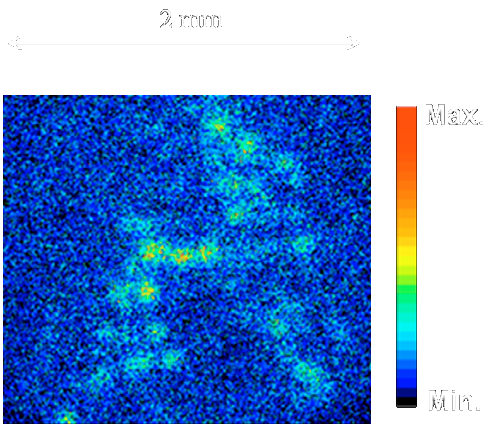

Figure 4. Map of Pb concentration on the surface of Figure 3. The highest values (brighter areas) match exactly the pattern of the stylus track as seen from the optical photograph.

This concentration in fact decreases in time from the death of the organism from which the find originates, with an exponential decrease characterised by a well known lifetime τ of about 8200 years (which corresponds to 5730 years for the so-called half-time $T_{1/2}$). Assuming as known also the initial concentration, the date of the find can be immediately obtained (the assumed initial value is not necessarily correct but the date thus obtained can be recalibrated).

The real problem is that the concentration to be measured is terribly small (one atom over thousand billions or even much less for an old sample), and in addition, in order to have a sufficient accuracy in the date, such a low concentration must be measured with great precision (1% uncertainty in measured concentration reflects into ± 80 years on radiocarbon date). Until about twenty-five years ago, to achieve such a precision and sensitivity, one needed to sacrifice a large mass of the find (tens or hundreds of grams). This was in fact the quantity of material necessary to achieve sufficient counting statistics in reasonable measuring times, using the β counting technique (according to the general radioactive decay law, the number of β decays of ^{14}C per unit time is proportional to the number of ^{14}C isotopes in the sample, so you can measure such decays and deduce ^{14}C concentration). Starting some twenty-five years ago, progressively an alternative method has been introduced, which is based on the direct measurements of the number of ^{14}C isotopes by mass spectrometry, using Tandem accelerators as ultra-sensitive, selective mass spectrometers. The great advantage of Accelerator Mass Spectrometry (AMS) is that the quantity of material needed for a dating is very low, in the order of milligrams. Dates back to 50000 years can be obtained, and for historical or pre-historical finds (back to 10000 years) the uncertainty on radiocarbon date can be kept within ± 40 years (the overall uncertainty on the date is often higher however, due to the recalibration procedure).

AMS has now almost totally ousted the traditional β counting method for radiocarbon dating. Presently, over forty laboratories in the world perform AMS to a larger or lesser extent, some of them being entirely dedicated. Over ten thousand dates per year are produced: the basic, hard, "obscure" work for the archaeologists.

In Europe, at least ten laboratories are active with AMS; many of them have a long experience, high qualification and a large throughput of dates per year. Recently, the possibility has been demonstrated of using even very compact tandems (< 1 MV terminal voltage) for precision (< 1%) ^{14}C dating.

In Italy, until three years ago no laboratories were equipped for routine AMS radiocarbon measurements. Today, three laboratories are active in this field (Naples and Lecce besides Florence). As mentioned above, the new 3 MV Tandetron of our LABEC laboratory is now fully operational, not only with IBA but also with AMS. We have also installed a well equipped sample preparation laboratory for radiocarbon measurements: indeed, the finds must be pre-treated, combusted and finally reduced to graphite before using them as pellets in the ion source of the Tandem where their ^{14}C concentration will be measured through AMS. The preparation procedure is very delicate because, due to the ultra-low concentrations of ^{14}C, any contamination with external carbon can modify significantly the final results.

For this reason, before we started with real dating campaigns, we spent months to check our reliability in the overall (preparation plus AMS) procedures needed to attain a radiocarbon date: in the first period of AMS operation, after accelerator commissioning was completed, only a large number of reproducibility tests with standards and background level measurements were indeed performed. Our ^{14}C background (i.e. the apparent amount detected in completely "dead", radiocarbon free, standards) corresponds to around 60000 years (AMS only), or 50000, also including contamination effects during sample preparation. This means that we are very competitive, at the highest level. Precision is also very good (< 0.5%, i.e. <± 40 y on radiocarbon age); the tests with standards of certified radiocarbon concentration are reproducible and satisfactory.

Following these reliability tests, a systematic activity of dating campaigns has started and about 200 dates of archaeological finds have been measured in the last few months. Among the first, let me quote studies aimed at authenticating some Medieval and Renaissance wood paintings, in collaboration with Opificio delle Pietre Dure in Florence; archaeo-metallurgical studies in northern Etruria, in collaboration with Sovrintendenza archeologica della Toscana, Department of Archaeology in Sienna, Department of Earth Science in Florence; dating of finds from excavations around Palazzo Vecchio and the Uffizi, in Florence, in collaboration with the Italian Ministry of Culture and the Department of Archaeology in Sienna. As I mentioned, more details on all our (old and new) activities can be found in our website, to which the reader is referred also for a list of references.

Acknowledgements

I wish to thank all the group of people working hard at LABEC: Luca Carraresi, Agnese Cartocci, Massimo Chiari, Mariaelena Fedi, Lorenzo Giuntini, Novella Grassi, Franco Lucarelli, Marco Manetti, Mirko Massi, Antonio Mirto, Silvia Nava, Francesco Taccetti. It is only thanks to them that the task of establishing the new lab in Florence and of having it so well operational in a short time has been successfully completed.

[1]As a suggested general reading for non-specialists wishing to learn about IBA applied to Cultural Heritage, I wish to mention a book published in the frame of a European COST action (G-1), i.e. *Applications of Ion Beam Analysis Techniques to Arts and Archaeometry, Eds.: M.A. Respaldiza and J. Gómez-Camacho, Universidad de Sevilla, 1997.*

X-RAY DIGITAL RADIOGRAPHY AND COMPUTED TOMOGRAPHY FOR CULTURAL HERITAGE

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Abstract

X-ray detection systems for high resolution Digital Radiography (DR) and Computed Tomography (CT) have been developed at the Physics Department of the University of Bologna. The research target is the development of systems to be applied in cultural heritage conservation and industrial radiology.

In the field of cultural heritage, different kind of objects (ancient necklaces, paintings, bronze or marble statues) have to be inspected in order to acquire significant information as the method used to assemble, the manufacturing techniques or the presence of defects. These features could be very useful, for example, for dating works of art or determining appropriate maintenance and restoration procedures. Among the advanced methods available, 3D CT can be successfully used for the investigation of ancient works of art because it preserves their integrity and provides images of inner parts, which are otherwise not visible.

KEYWORDS: COMPUTED TOMOGRAPHY, CULTURAL HERITAGE, DIGITAL RADIOGRAPHY, MICRO-TOMOGRAPHY

KULCSSZAVAK: SZÁMÍTÓGÉPES TOMOGRÁFIA, KULTURÁLIS ÖRÖKSÉG, DIGITÁLIS RADIOGRÁFIA, MIKRO-TOMOGRÁFIA

Introduction

Several high-resolution CT systems have been developed to investigate objects of different sizes (from micro to macro) at the Physics Department. For example, we have carried out the micro CT reconstruction of Roman human tooth with dental caries (found in the “Isola Sacra” necropolis); as well as the cone beam CT analysis on an Egyptian cat-shaped coffin exhibiting the inner mummy; up to the CT study of an large ancient globe (2 m of diameter). This globe was created by a Dominican monk, Egnazio Danti, around 1567 and is located in Palazzo Vecchio, at Florence. The very high resolution reached investigating small objects is an important result other than tomography on a big object, like the globe, is an absolute innovation. A 3D CT investigation is being in project to determine how much deterioration has occurred on the ankles of David, the towering marble figure sculpted by Michelangelo, a very exciting purpose that we will achieve in collaboration with Lawrence Livermore National Laboratory.

A new linear array detector for high resolutions and low dose digital radiography for painting was realised. This new instrument is able to acquire radiological image with an amount of dose one hundred times reduced less than standard film. The system was tested on a benchmark panel with some pigments provided by the "Opificio delle Pietre Dure" (OPD) a well known Restoration Centre in Florence. The resolution and the image contrast reached by the scanning system were superior to that of the common film systems used at the Institute. Moreover, in collaboration with the National Gallery of Bologna and OPD, was performed an X-ray investigation of the inner structure of two small painted "tablets", made of wood, and recognised as an artwork of Gentile da Fabriano, an important painter of the XIII century. Different techniques were used: conventional film radiography, digital radiography and computed tomography, the latter two with innovative equipment of the Department of Physics in Bologna.

Material and Methods

Methods of diagnosis based firstly on X-ray digital radiography (DR) and then on computed tomography (CT), are more and more used for the conservation and restoration in the cultural heritage field. This kind of analysis can help also to understand the construction techniques and the “history” of the object under examination (Casali 2006).

As the size of objects of cultural interest varies greatly, from small fragments to large works of art, it is necessary to develop measurement systems for each typology of objects. For small objects (i.e. fossil teeth and ancient jewels) it is necessary to use high spatial resolution detectors, for big or thick objects it is necessary to use very efficient detection systems. The detectors developed and used, at the Physic Department the University of Bologna, can be one dimensional (linear detectors) as well as two dimensional (planar detectors).

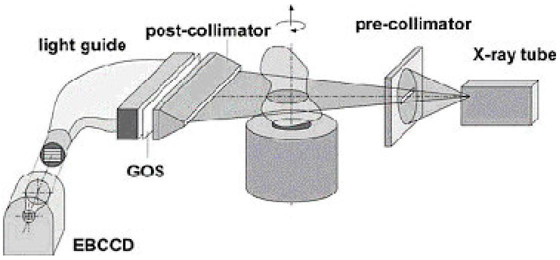

Figure 1.

Scheme of the linear detector set-up

A scheme of the linear detector set-up is shown in **Fig. 1.** The device consists in an X-ray generator, a pre-collimator, a mechanical translation stage, a post-collimator and a strip of Gd_2O_2S:Tb. This scintillator is optically coupled with the photocathode of an EBCCD camera through a coherent image light guide made of thin glass fibres. The most new feature of the system (patented by the University of Bologna) is the ‘‘optical guide-EBCCD camera’’ combination. The input end of the optical guide, at the head of the detector, is 129 mm wide and 1.45 mm high. The output end of the optical guide has approximately a square shape and fits into the 1 in. diameter photocathode of the EBCCD camera. The guide is composed of seven 18.4 × 1.45 mm^2 bundles, about 50 cm long. Each bundle transports the light in a coherent way to preserve spatial information. The EBCCD camera is provided with a high voltage tube as in a conventional image intensifier. The difference is that electrons hit directly the substrate of the CCD that is sealed inside the tube. In this way a higher conversion efficiency and a higher gain (up to 2000) are obtained. Moreover, the EBCDD camera is compact and small with respect to a conventional image intensifier (Bettuzzi et al. 2004).

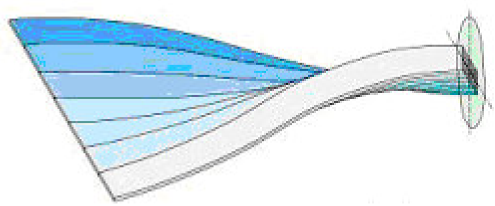

Figure 2.

Rectangular-to-linear fibre optic adapter: the linear input face is converted on a rectangular exit

Figure 2 shows a picture of the fibre optic adapter. It is made of glass microfibres, each one with a diameter of about 20 μm. The microscopic size of the fibres provides a great flexibility to the adapter. The bundles are aligned on the input face, by thus allowing a linear scanning geometry. On the other hand, the fibre-optic bundles are stacked at the exit, in order to fit the shape of the photocathode of the EBCCD camera.

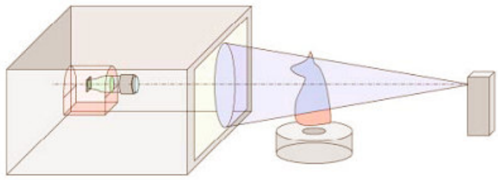

Figure 3.

Diagram of the intensified TC system

A diagram of an example of the two-dimensional detection system configuration is shown in **Figure 3**. A 30×40 cm^2 GOS screen is mounted on the entrance window of a large box that both acts as a support and provides proper shielding for the detection system. By changing the distance between the EBCCD and the scintillating screen and adopting a suitable lens on the photocathode, it is possible to obtain high resolution images of a smaller area and a radiographic zoom of a specific zone. The set-up is similar to that of the micro-CT system, but this intensified system permits investigation of bigger objects with a resolution of about 200 μm (depending on the magnification factor, (Pasini et al. 2004)).

Moreover the characteristics of the X-ray source are equally important. In fact, for doing DR or CT with spatial resolution of the order of 10 – 20 mm, it is necessary to use the so-called microfocus, having focal spot not larger than 5 mm. Recently more sophisticated X-ray tubes have been developed having focal spots not larger than 1 mm (nanofocus).

The X-ray sources of interest can be summarised thus:

- X-ray tubes (from 5 kV to 450 kV) with the versions micro- and nanofocus;

- linear accelerators (from 2 to 15 MV);

- synchrotron light (from 5 keV to 100 keV).

For light energy X-ray CT we have used a 9MV linear accelerator of Aviogroup-Rome. With that source we did a CT of an old Roman jug with bronze coins (**Fig. 4.**).

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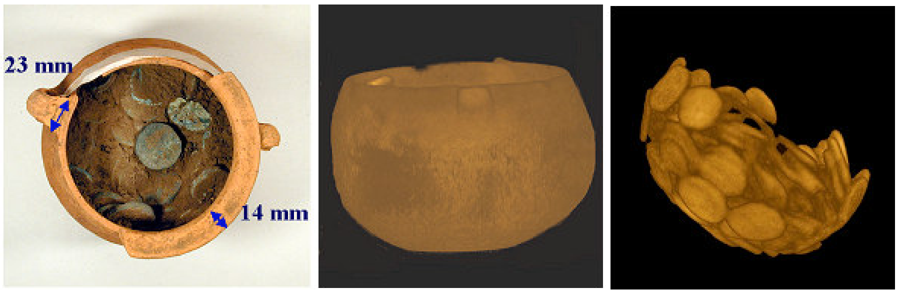

Figure 4.

Picture of the old Roman jug with bronze coins (left), CT images showing the jug and the inner coins (right)

Results

In the last few years at the Physics Department of the University of Bologna, X-ray detection systems for high resolution DR and CT have been developed and successfully employed for the analysis of samples of different size and composition. For example, in the framework of a collaboration between and the Archaeological Museum of Bologna and the Physics Department, important archaeological findings and works of art have been investigated by means of Digital Radiography and Computed Tomography. In particular, these methods have been used to inspect bronze object of the Etruscan section (Rossi et al. 1999, **Fig. 5**) and small mummies from the Egyptian Collection (**Fig. 6**).

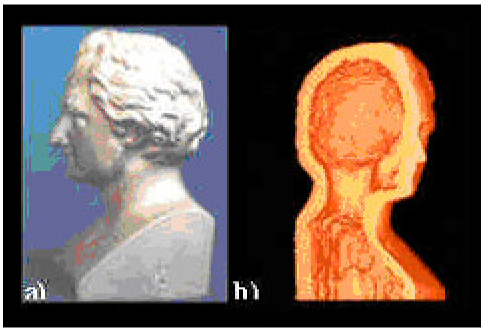

Figure 5.

a) Small Etruscan bronze head, b) 3D Computed Tomography

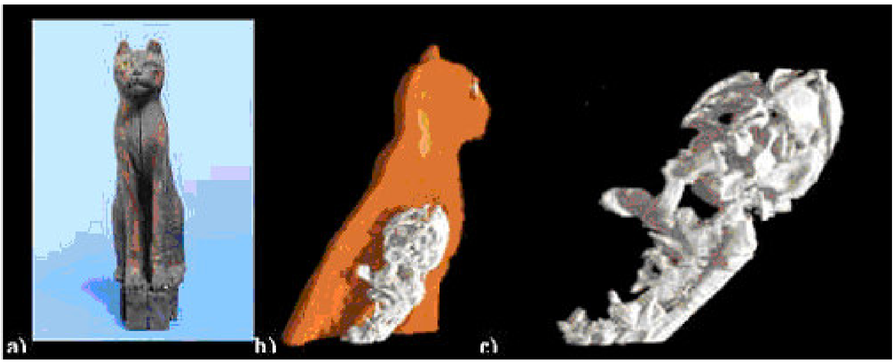

Figure 6.

a) Egyptian cat-shaped coffin with a cat mummy included inside,

b) 3D Computed Tomography,

c) zoom of internal skeleton.

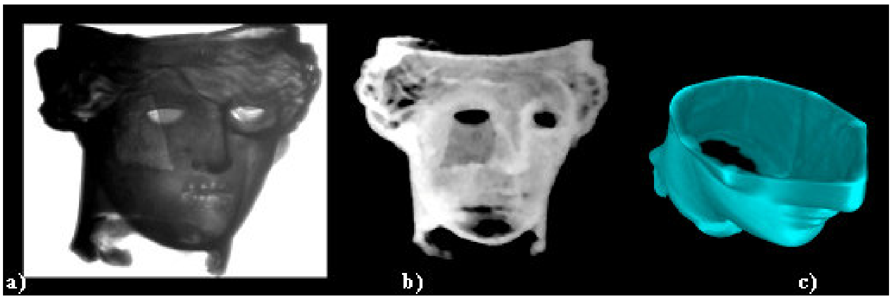

Figure 7.

a) Greek Bronze Head

b) Digital Radiography

c) 3D elaboration

Other investigations have been carried out in collaboration with the Getty Conservation Institute (GCI) of Los Angeles (**Fig. 7**) using an X-ray tube up to 450 kV. With GCI is going to start a research program focused on CT of an ancient bronze statue for trying to understand the welding technique of Romans and Greeks.

In the frame of a collaboration between OPD and the National Gallery of Bologna, a comparison has been performed between the digital radiography (Rossi et al. 2000) and the traditional radiography with films, with the conclusion that the DR has the same resolution as the traditional one but with much less radiation dose released to the painting (**Fig. 8**).

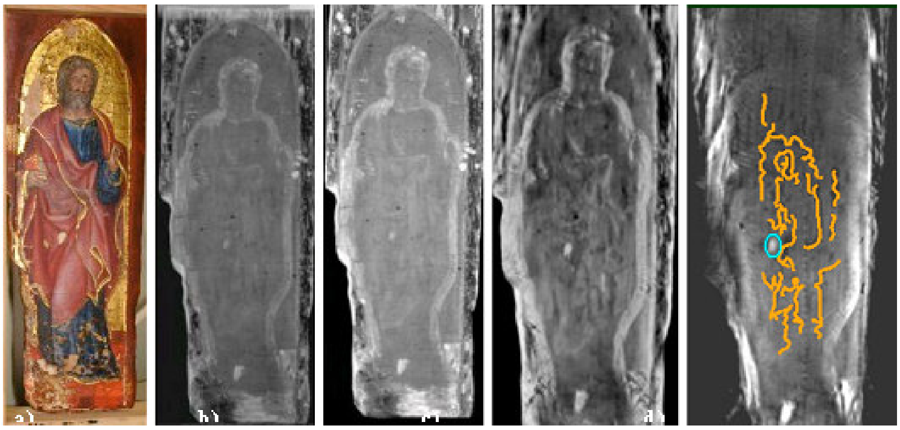

Figure 8.

Painted "tablets", artwork of Gentile da Fabriano:

a) Picture,

b) Conventional film radiography,

c) Digital Radiography with our System,

d) Digital Tomography with our System (section)

e) Warm holes and pipes, CT section.

The developed systems have a strong sensitivity to low flux X-ray radiation. For this reason it is possible to obtain a significant reduction of the absorbed dose.

Among the examined works of art, it is worth mentioning the big globe built in 1567 by the Dominican monk Egnazio Danti and located in Palazzo Vecchio, at Florence, Italy (**Fig. 9/a**). A restoration project is ongoing for returning the globe to its original magnificence; within this project, a CT of the globe was achieved, for exploring the nature and the conditions of the inner structure. The main problem of getting a complete CT was related to the large size of this masterpiece (220 cm in diameter) and to the need of achieving an in situ analysis in a museum with a lot of visitors. For these reasons an ad hoc experimental apparatus was realised and set-up at Palazzo Vecchio. The 3D CT reconstruction of the globe has clearly shown the entire inner structure that was never seen before (**Fig. 9/b**, Casali et al. 2005).

Conclusions

The digital radiography and computed tomography are two new and interesting fields of non-destructive evaluations and a tools for scientific investigations.

For example, the 3D CT reconstruction of the globe clearly shows the entire inner structure, how it was deformed during time, how it could be restored. All the inner structure, made of iron with a total weight of about 350 kg, was estimated from the segmented 3D reconstruction. Another important application is the study of the metal fusion technique used by the ancient artists (Greeks, Etruscans and Romans). Moreover the 3D CT image can help the restores in designing the restoration and conservation of ancient bronze statues.

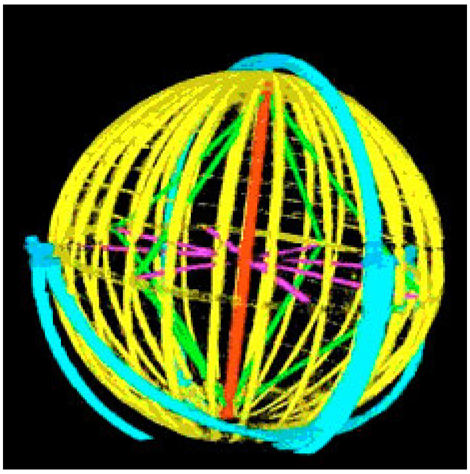

Figure 9.

a) The big globe built in 1567 by the Dominican monk Egnazio Danti and located in Palazzo Vecchio, at Florence, Italy (left);

b) The 3D inner structure reconstruction of Danti’s globe (right).

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It should however be pointed out that the CT technique is difficult and expensive. In fact, for having good CT images, many hundred of radiographies are necessary with the use of very expensive equipment for moving the objects with a precision of a few microns (Rossi et al. 2002, Rossi et al. 2004).

The easiness with which CT can be performed in the medical field may be misleading: medical CT was optimised for the human body (composed mainly of water) and cannot be successfully used on bodies with different density. In order to perform good, non-destructive evaluations, the most suitable DR or CT system (source, moving equipment, detector and elaboration software) must be carefully chosen to avoid obtaining meaningless results (Casali et al. 2003, Casali 2006).

Acknowledgements

The Author wishes to thank all his young contributors, pictured in **Figure 11** (Matteo Bettuzzi, Davide Bianconi, Rosa Brancaccio, Samantha Cornacchia, Carlotta Cucchi, Emilia Di Nicola, Alessandro Fabbri, Marilisa Giordano, Nico Lanconelli, Alice Miceli, Maria Pia Morigi, Alessandro Pasini, Davide Romani, Alberto Rossi), for their enthusiasm in applying advanced techniques in the interest of the conservation and understanding important cultural treasures. Heartfelt thanks to Serena Pini (Musei Comunali, Florence), to Giacomo Chiari, Senior Scientist of the Getty Conservation Institute, to Alfredo Aldrovandi of the Opificio delle Pietre Dure, for their very appreciated collaboration.

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POSSIBILITIES AND LIMITATIONS IN THE ARCHAEOGENETIC ANALYSIS OF ANCIENT HUMAN REMAINS

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Abstract

Archaeogenetic investigations – parallel to the wide expansion of molecular genetics – have recently gained importance in archaeology and population history. This positive change in its role in historical research is based partly on the technological development of the last two decades and partly on the recognition of the fundamental conservatism of the DNA.

The field was open for a multi-respect analysis of the DNA of several thousand years old human remains. The polymorphism of the DNA, especially on certain sections of the mitochondrial DNA, offered the possibility for the most thorough examination ever in relation to the spread and genetic variability of the human species. Because of the fragmentary character of the preserved ancient DNA sections, the morphometric features of the human skeleton and the genetic haplogroups formed by the DNA-based polymorphism cannot be correlated. In fortunate cases, archaeogenetic investigations make it possible to study illnesses of genetic origin or analyse kinship relations in smaller burial groups. The examination of patrilineal and autosomatic inheritance can be of great help in answering the major questions of the population history of the Carpathian Basin.

The most recent investigations concentrate on the testing of archaeological and historical preconceptions regarding the eighth to twelfth centuries, with special emphasis on the problem of population and ethnic group. The most spectacular results, however, can be expected in relation to the population problems of the Neolithic. The archaeogenetic laboratory in the Archaeological Institute of the Hungarian Academy of Sciences was established to answer these questions.

The long-term financing of such investigations, however, has not been solved properly, not least because of the time needed and the difficulties one has to face when trying to provide an interdisciplinary interpretation. In order to decrease the possibility of modern human DNA contamination, there is a recent tendency to limit the number of research groups and focus the limited resources – among them the available grants and funds – in the major archaeogenetic research centres.

KEYWORDS: ANCIENT DNA, MITOCHONDRIAL POLYMORPHISM, CONTAMINATION, ARCHAEOGENETIC RESEARCH

KULCSSZAVAK: DNA, MITOKONDRIÁLIS POLIMORFIZMUS, SZENNYEZŐDÉS, ARCHEOGENETIKAI KUTATÁS

Introduction

During the second half of the last century it became a fundamental paradigm that answering certain questions requires a close co-operation between various branches of research. Among humanities, one of the most imminently affected discipline was archaeology, that embraced a host of scientific methods in addition to its traditional auxiliary or sub-disciplines, such as physical anthropology, pedology and geology, archaeobotany and archaeozoology. A range of material studies and various dating methods were introduced into archaeological research that have formed a new trend. By the 1960s and 1970s, „New Archaeology” gained a decisive role in archaeological research in the „Western World”. The interpretation and proportion of results yielded by natural sciences, however, were often misrepresented in the conclusions drawn by archaeologists and historians due to their schooling, approach and the insufficient knowledge of the

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faults and limitations of the methods. In hindsight, it may be considered inevitable that trust in the direct application of scientific results as panacea for archaeological problems eventually dissipated. As a predictable backlash, strong criticism and a sceptical attitude toward results gained by using new scientific methods became commonplace in many historical-archaeological treatises. The application of scientific dating methods suffered most from this criticism, although classical physical anthropology, trying to rejuvenate itself by the extensive use of biostatistics, has also been bruised.

Revolutionary changes that took place in molecular biology and biotechnology from the 1970s onwards, have lent a new momentum to interdisciplinary research in archaeology. The most evident sign of this was that archaeogenetics, that has since developed into a new, independent field of research, was included into archaeological investigations. The appearance of molecular genetic profiling may be traced back to three main reasons. These were as follows:

- the increasingly precise understanding of the function and structure of DNA, that lead to the recognition of its fundamental conservatism,
- the development and availability of the polymerase chain reaction (PCR) technology,
- the recognition of DNA preservation capacity of bones from archaeological contexts.

The emergence and rapid proliferation of archaeogenetic investigations in international scientific research are largely the result of the more-or-less simultaneous application of DNA based methods using archaic human and animal remains, and research in forensic medicine, criminology and various areas of biotechnology. Results of global importance have been born in the shadow of the much publicised human genome project (HUGO), stem cell research and tumour genetics. These included determining the complete base sequence of mtDNA and the description of polymorph characteristics of the non-coding region, as well as the identification of the genetic and geographical origins of the human species, which helped refute the theory of multiregional evolution. The maternal and paternal lines of inheritance could be mapped, the tree of haplogroups was drawn and the process of settling the various continents could be clarified. Meanwhile, a consensus was reached in professional circles as to the technology and laboratory environment required for the isolation and amplification of ancient DNA stock. An increasing number of research teams started publishing archaic mtDNA sequences, and experience acquired in other areas of genomics have also been successfully applied. These included the criteria for grouping, the methods of establishing chronological sequences for mutations as well as the clarification of origins of and relationships between living human populations. As of today, sufficient knowledge has been accumulated to state that the successful application of DNA studies is limited to at most ten thousand years. It seems, however, that within this time interval only the remains of people buried during the last two or three millennia can be studied on a regular basis, in spite of the fact that the DNA preservation and consequent analytical potential of bones is less dependent on the absolute time of deposition than on the taphonomic effects of micro-environmental factors in the deposit.

Studying archaic DNA samples has had its own childhood diseases. The most important of these has been the problem of contamination by modern human DNA. Strict laboratory protocols have been developed in order to exclude false positive results. These include the complete spatial isolation of the pre-PCR and post-PCR phases of processing, as well as the parallel analysis of samples. In recent years, cloning has gained increasing importance in identifying contamination by modern DNA. Just for general information: if the DNA content of an archaic bone sample recovered under average circumstances is taken as one unit (1, in a mathematical sense), the number of copies obtained after amplification may be on the order of millions or even tens of millions (10^{6-7}). This order of magnitude corresponds to the concentration found in the fresh, live tissue. However, recently unrealistic requirements have been put forward in connection with cloning to the detriment of the publication possibilities of small laboratories and research teams. This is particularly worth mentioning here, since there is a general consensus that DNA obtainable from fossil bone remains tends to be heavily fragmented. The size of such fragments does not exceed a few hundred pairs of bases. Should it be possible to amplify segments longer than this, one should always be aware of the risk of contamination.

Archaeogenetic research in the Carpathian Basin

Archaeological investigations in the Carpathian Basin have built their chronologies on theoretically assumed population changes, reconstructed on the basis of material remains and written sources. Since the precision of the latter in reconstructing events in population history is limited, the methods of archaeogenetic investigations complementing the results of traditional physical anthropological studies are of prominent importance. In order to facilitate complementary research carried out in this field, a research agreement was signed in 2001 between the Archaeological Institute of the

Hungarian Academy of Sciences and the Institute of Genetics, Biological Research Center (Szeged). This co-operation has been supported by state-sponsored grants. In order to increase the efficiency of research work and to create a database, a PCR laboratory was created in the Archaeological Institute of the Hungarian Academy of Sciences.

The construction of this laboratory took place between 2002 and 2004. The design of working areas was strictly defined by the protocol requirements of isolating and typing archaic DNA. The processes of isolation and the PCR phase had to be separated as much as possible. Therefore, the so-called preparation room, used in the isolation of archaic DNA and the PCR processing area are located at the opposite ends of a corridor, with a general preparation room in between. An isolated space within this latter room is maintained for polishing and pulverising ancient human bone.

Our investigations, carried out in co-operation with the research team of the Biological Research Center (BRC) have shown that the mtDNA sequences identified in the AD 10th century samples belong to Asiatic haplotype groups in a far greater proportion than those taken from modern Hungarian populations. Currently, our data base contains the sequences of 70 archaic samples drawn from 120 graves representing the 8-12th century populations of the Carpathian Basin. Recently, our research has targeted the amplification of our data base both in time and space, the mapping of the paternal lines of inheritance and the testing of the relevance of using autosomal markers in our studies. In addition to human bone samples, the analysis of animal remains has also been carried out in the BRC. To date, mtDNA sequences have been obtained for horse, cattle and sheep.

There is, however, a period in the prehistory of the Carpathian Basin which is of essential significance from the viewpoint of the population history of the entire continent of Europe: the period of the Early and Middle Neolithic. Investigating the spread and directions of Neolithization using methods of archaeogenetics is one of the hottest research subjects in European archaeology. From this point of view, it is of special interest that, on the maternal side, the overwhelming majority of populations inhabiting modern Europe originates from people who lived in this area already in Palaeolithic times. Only one of the seven main haplogroups characteristic of modern European populations is of Neolithic origins. The basic question of how this Neolithic group spread and exerted its population genetic effects can and should be studied in the area bordered by the northern Balkans, the Alps and the Carpathian Mountains.

Limitations of archaeogenetic analysis

As mentioned before, the application of scientific results in the interpretation of historical processes is hampered by major difficulties. It is therefore necessary to face the limitations which *ab ovo* determine the direct applicability of results obtained by archaeogenetic research. These difficulties may be summarised as follows:

- limitations on conclusions and difficulties of interpretation and chronology;
- limitations posed by the “inaccuracy” of databases;
- limitations diachronic and taphonomic of DNA preservation, depending on the microenvironment;
- limitations of technology and financing research.

The difficulties of interpretation are mainly due to the fact that samples subjected to genetic analysis already represent an archaeological-historical, consequently also chronological, preconception. Deciding, who among the AD 10th century population should be considered first generation, “conquering” Hungarian, is currently determined by social scientific methods. However, bone samples taken from graves or sets of burials selected and characterised from an archaeological point of view, by definition “absorb” uncertainties of these selection criteria. Experts in genetics and biostatistics will treat them as unambiguous raw data.

When evaluating our results, it is also important to consider the relationships between the databases at our disposal. Such databases shall be evaluated according to geographical and chronological aspects. One may say in general terms that most reference databases are built on “modern” samples. This means that the information concerning the population of a given area either has little or no time depth, or offers possibilities of interpretation of extremely long time spans, as is the case with mtDNA haplogroups. This is clearly exemplified by the comparison between modern Hungarian “samples” and populations inhabiting geographical areas considered to be of outstanding importance from the viewpoint of ancient Hungarian history (regardless of blood typing or other genetic characters). This solution is understandable, considering the scarcity of relevant data representing this historical and archaeological period. On the other hand, one cannot ignore the fact that populations included in this comparison also have their own “prehistories” with their own chronological and spatial dimensions. In other words, the sample used in comparisons can be considered “constant” only in exceptional cases.

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Even using the ample evidence of historical, archaeological and linguistic data, populations cannot be characterised as precisely as would be necessary to support the conclusions drawn from them.

Similarities and differences between the patterns identified on the basis of the studied genetic traits may be analysed descriptively adhering to certain rules. It is a fundamental problem, however, that our possibilities are extremely limited in answering basic questions such as the sources and especially the timing of the differences between populations. The starting point of our investigations has been that the differences between the Asiatic haplotype distributions in AD 10th century populations in the Carpathian Basin and modern Hungarian populations was the effect of the 10th century conquering Hungarians of Asiatic origin. However, in order to correctly evaluate the changes of the haplotype patterning of the populations within this geographical area, one must also consider the possibility of earlier and later occurrences of Asiatic genetic elements. We should not forget that our present investigations have not been able to prove the exclusively AD 10th century origin of the Asiatic character of the maternal line of modern Hungarians. Data representing several consecutive phases are required to eventually understand the processes of change and draw realistic conclusions concerning the processes of population history.

The scientific success of this research is strongly influenced by a special contradiction. Natural scientists are under constant pressure to produce and publish new ideas, while historical studies require the accumulation of databases of representative sizes, made possible only by long term investigations. Frequently, this contradiction is not as much the product of differing disciplines and their differing paradigms, but rather of incompatible financing strategies and often the lack of appropriate research plans. Molecular genetics undoubtedly needs the freedom to interpret its results within its own frame of reference. This can be achieved by analysing a relatively small number of samples. Drawing historical conclusions and reconstructing the underlying processes, however, require larger series. Should an outlier occur within the mtDNS patterning of a certain time period, it is not sufficient to interpret it as a consequence of a genetic anomaly. It is for this reason that the division of research between our archaeogenetic laboratory and the research team based in Szeged has resulted in a fruitful co-operation. The development of methods and study of primers has been taking place in the Institute of Genetics, while the Archaeological Institute is in charge of studies of historical relevance that require the time consuming creation of major databases.

One should not underestimate the significance of sampling problems either. This is especially important when we do not have an opportunity to select the best of several samples. To date, research has shown that the relationship between the DNA preservation potential of bones (and the related success of analysis) and time of deposition is not as important as the effects of micro- and macro-environmental factors, burial rite and isolation that all influence the success of the PCR reaction. For example, bone remains of the Sarmatian population are notoriously poorly preserved in the Carpathian Basin, therefore little is known of the morphological traits of these people. It is possible that the molecular genetic profile of this population will be similarly difficult to establish to an extent that could meaningfully contribute to answering questions of population history. It is especially frustrating when sampling bones from burials of outstanding chronological importance turns out to be unsuccessful. It is also problematic to evaluate the remains of children. In the case of juvenile skeletons, which already contain only a negligible amount of cortical bone, it is more difficult to take non-contaminated samples of reasonable quantity to begin with. This has a fundamental bearing upon the success of isolation and amplification. Meanwhile, in order to clarify internal kinship relations and chronological sequences within groups of burials, one should know their genetic affiliations as well. An additional difficulty is posed by the unfortunate fact that many physical anthropologists are hesitant when destructive sampling techniques must be used, even on a relatively small scale. This is in spite of the fact that a consensus has been developing in the international literature concerning the quantitative and technical standards of proper sampling.

Hopefully, in addition to the critical comments put forward in this presentation, I could also direct your attention to the actual importance of archaeogenetic research itself. As far as I am concerned, I consider it a special honour and great luck that, as an historian trained in biological anthropology, I have had the opportunity to witness and actively promote the establishment of this typically interdisciplinary project in an archaeological institution.

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LASER TECHNIQUES FOR CONSERVATION OF ARTWORKS

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Abstract

The potential of laser techniques in conservation has needed a long development period to be fully demonstrated. The possibility to achieve a very precise and selective removal of deteriorated materials was implemented through a series of interdisciplinary studies focussed on laser ablation of specific materials. A suitable choice of the laser types and of the operating parameters could optimise the cleaning results, avoiding side effects while preserving the historical layers behind deposits and encrustation. An extensive validation carried out on a number of renowned masterpieces has definitely spread the interest of the conservation community for laser techniques in many European countries. The paper reviews the development of specific laser cleaning techniques for stone, metals, pigments and organic substances, which require different choices of laser wavelength and pulse width.

KEYWORDS: LASER TECHNOLOGY, CONSERVATION

KULCSSZAVAK: LÉZER TECHNOLÓGIA, RESTAURÁLÁS

Introduction

The remnants of past civilisations are an important part of the historical and cultural identity of the population of each country. The uniqueness of each piece of this treasury justifies the need of the most developed means in order to preserve the material itself against the many sources of deterioration. Because of this, the conservation community has always explored the potential of newly developed science and technology for solving the problems they are everyday facing. Since the opening of a modern meaning of restoration by Brandi (1963), chemistry has been mostly involved, providing reactants, poultices for consolidation and cleaning, coatings for protection. Physics has also given very important contributions for diagnostic methods such as microscopy, optical and X-ray investigations. Laser techniques came in about more than thirty years ago (Lazzerini & Asmus 1973), giving immediately very promising results not only for diagnostic but also for restoration procedures. The potential application for the delicate phase of cleaning was immediately understood but the development of successful laser techniques in conservation took necessarily many years. The aim of this paper is to present a review of laser techniques in conservation, focussing on the most important application in the cleaning phase of a restoration intervention. They have been validated in a significant number of case studies and today have demonstrated to provide advanced solutions for cleaning problems that allow a level of precision and control unreachable by other methods.

Laser cleaning of stone

Stones exposed to urban environments develop on the surface the typical sulphation process, with calcium carbonate turned in a gypsum matrix, enclosing carbon particles deposits, finally leading to a weakening of the material stability and in the worst case to a loss of materials. In this case the restoration intervention becomes often an urgent need, in order to stabilise the state of conservation of the materials. For the delicate phase of the degraded material removal the intervention technique should be able to provide a minimal invasive action, with the possibility to being selective between the encrustation and the historical layers. Traditional cleaning methodologies usually employ mechanical removal or chemical reactions. The erosion of the encrustation by sand-blasting is caused by the kinetic energy and cannot be selective. Chemical perfusion with Ammonium carbonate or other chemical reactants have drawbacks, with scarce control on the effects. Laser cleaning is a good candidate for this task (**Fig. 1**), being potentially very progressive, precise and selective. The physical process involved is laser ablation of inorganic (sometimes also organic) layers composing the deterioration crust. The most important characteristics are:

Laser ablation takes place when a pulse of laser radiation is absorbed at the surface of a material, determining a sudden transition of its solid phase to another phase (gas, vapour, plasma) if the energy density overcomes a certain level defined as the threshold.

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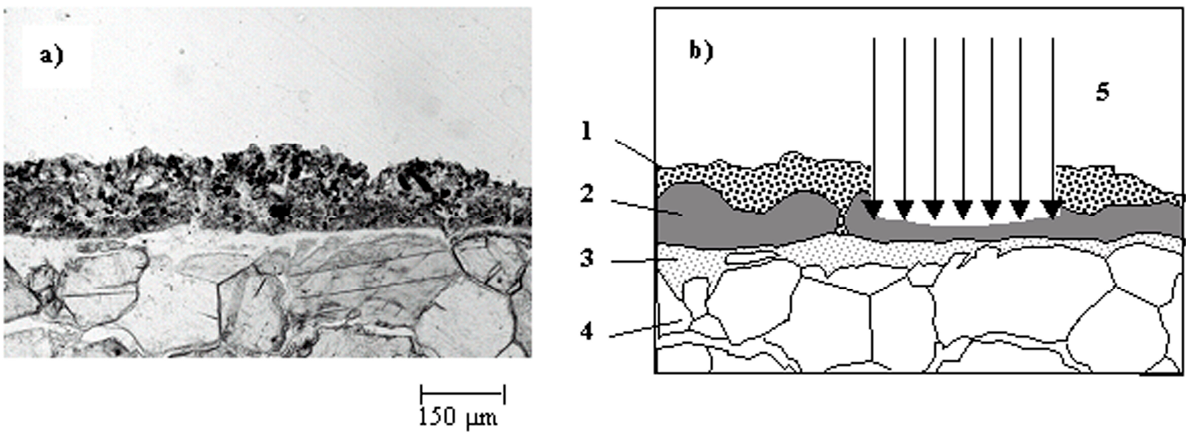

Figure 1.

a) Example of stratigraphy of a deteriorated marble observed by an ultra thin section.

b) Descriptive scheme: 1) black crust, 2) sulphated Ca-oxalates film (showing craquelure), 3) surface pseudomorphic sulphation layer i.e. reproducing the shape of the original surface, 4) calcite crystals with intergranular decohesion, 5) laser cleaning proceeds in a controlled way down to the oxalate layer.

The sudden phase transition is mainly due to photo-thermal effects rising the temperature of the material up to a hot vapour that expands quickly in the surroundings, producing a material removal.

Laser cleaning is essentially a surface treatment, where only a thin layer limited to a few microns or less than a micron is directly involved by absorption of light, while chemical methods usually perfuse with solvents internal layers without control;

The ablation threshold of high absorption materials (black encrustation for example) is lower than low absorption materials (light colour stone for example), so that a selective removal is possible without problems to preserve historical layers, as it is shown in **Fig. 2**.

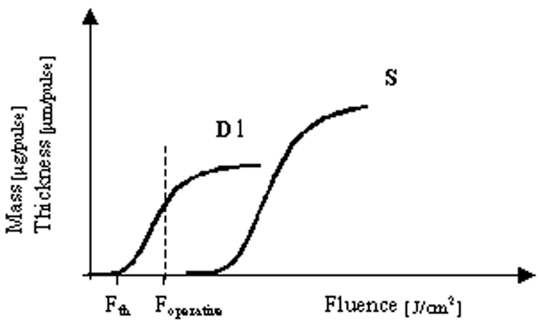

Figure 2.

Typical laser ablation curves for deposit (D) and substrate (S). They describe the dependence of the mass or thickness removed by the laser ablation process by varying the fluence or energy density. Setting the laser irradiation at the fluence value $F_{operative}$ an effective removal of deposit takes place, while no removal of the substrate is possible.

Accurate control of the treated volume is possible, due to the very progressive action of the laser, removing the encrustation pulse after pulse.

The applications of laser cleaning to stone materials are certainly the most advanced ones. For this problem is now recognised that laser cleaning represents the best possible technique, resulting selective and progressive. Nevertheless many studies and experimentation were needed to arrive at the present point after the first pioneering test by Lazzerini & Asmus in Venice in the 1972 (using ruby lasers). Since then the most practical Nd:YAG lasers emitting in the infrared spectrum (IR) at 1.06 μm, operating in Q-switch mode with very short (about 5-10 nS) and intense pulses have been employed if the following experiments. In Italy several laser cleaning tests were carried out in the ‘70s and in the ‘80s in Venice, Padua, Cremona by Calcagno (1987). The interest in laser cleaning started again in the ‘90s in France by Orial (1989) with these lasers employed for the restoration of the portals of cathedrals in Amiens, Mantes-La Jolie, Paris, Chartres, Saint Denis etc. Another group (Cooper et al. 1995) was studying laser cleaning of artefacts in Liverpool using Nd:YAG lasers, while in Crete Fotakis (1995) began to consider also the category of excimer laser (a gas laser emitting in the ultraviolet spectrum) for icons and paintings, and in Germany Olainek et al. (1997) considered also the problems of medieval glass as candidates for excimer laser cleaning.

In Vienna Calcagno, Nimmrichter et al. (1997) could use laser cleaning on a large scale at the restoration of the St. Stephan cathedral, producing another important step in the validation of the laser technique.

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Nevertheless these investigations remained somehow isolated without a real spread of laser techniques for the conservation of stone. This was partially due to side effects shown after laser cleaning such as yellowing or discoloration of the stone. This phenomenon observed by several authors has had different interpretations and perception. In Greece using the term discoloration is underlined that the original colour of the historical patina is modified. In France the yellowing (Labourè et al 2000) shown by cleaning of limestone with Q-switch Nd:YAG laser was simply softened by washing gently the surface, demonstrating that it was due to a superficial fall of micro particles after the ablation. In Germany SEM analysis has shown Fe nanoparticles after ablation that could justify the yellowing.

In order to overcome the limits shown by Q-switch mode operation, a special Nd:YAG laser operating with long and less intense pulses in the microsecond range was developed by Siano et al. (1997) in Florence. This new laser avoids yellowing effects and has been successfully employed in the restoration yard of the church of San Frediano in Pisa, at the *Porta della Mandorla* by Nanni di Banco, the north door of the Santa Maria del Fiore Cathedral in Florence, at the *Porta di San Ranieri* at the Duomo of Pisa, on many fragments of the original *Fonte Gaia* by Jacopo della Quercia, formerly set in *Piazza del Campo* a Siena. In all these restoration activity the microsecond laser technique has allowed to discriminate the proper layers removal in very complex stratigraphy overimposing various layers of pigmentation, sulphation, oxalate and so on.

Important applications of the laser techniques have been carried out on marble statues severely degraded by exposition for centuries to the sulphation process. Several masterpieces by Donatello such as the *Prophet Habacuc*, shown in **Fig. 3**. during the restoration, the *Pulpit* in the Prato Cathedral and Nanni di Banco's *Santi Quattro Coronati* have been successfully restored using laser cleaning especially when other traditional techniques such as micro sandblasting or chemical treatment could not ensure the achievement of the result. This has been the case of gilding traces, left by the action of time in the dresses borders and in the hair. Only the delicate calibration possible with the microsecond pulses laser cleaning has allowed preserving them at best.

In Greece, archaeological pieces such as a marble statue of *Hermes*, and recently a panel of the west frieze of the *Parthenon* were cleaned using a Q-switch Nd:YAG laser and combining the emissions at both the fundamental wavelength 1.06 (IR) and at the third harmonic (UV).

Figure 3.

The marble statue of the *Prophet Habacuc* during laser cleaning at the *Opificio delle Pietre Dure* in Florence. The restorer employs the hand piece as a tool on the statue.

The combination of IR and UV radiation could avoid yellowing of the cleaned surface, which was observed using IR emission alone.

Detailed descriptions of these case studies are reported in the proceedings of the International Conference LACONA (Lasers in the Conservation of Artworks) edited by Salimbeni R. & Bonsanti G. (2000), Verges-Belmin V. (2003), Dickmann K., Fotakis C. & Asmus J.F. (2005). The COST Action G7 provides a reference site on the web for this topic at http://alpha1.infim.ro/cost/.

The two regimes (Q-switch and Short Free Running) have also different features in respect of the beam delivery:

Laser systems emitting Q-switch high intensity pulses employ typically articulated arms (with mirrors at each turning joint) to propagate the beam to a hand-piece, because optical fibres could be damaged by the beam energy density higher than the damage threshold of silica fibres. Consequently Q-switch lasers may give a maximum pulse energy in the order of 300 mJ out of 1 mm diameter fibre, or higher pulse energy (up to 1 J) using an articulated arm. The use of a laser equipped with an articulated arm is practical only in a laboratory condition, where the relative positioning of the laser in respect of the artwork is easy. For outdoor use on scaffoldings the use of Q-switch lasers have given problems for the weight, the positioning and for the possibility of heavy environmental conditions such as rain, humidity and dust.

Short Free Running Lasers emitting microsecond pulses may couple easily 2 J in a silica fibre, without risks of damage, consequently they are equipped with optical cables of various lengths, to operate on the scaffoldings keeping the laser instrument in a safe place against the environmental danger, as shown in **Fig. 4** for the restoration of the Mausoleum of Theodoric in Ravenna. Using the fibre the hand-piece provides a quite homogeneous spot, which is a premise of a well controlled material removal all over the irradiated area.

Nowadays all the spread use of lasers in conservation of stone is based on the clear task to provide a very precise cleaning where the highly valuable decorations (statues, relieves, friezes, columns capitols, coats of arms, labels etc.) are worth of the best possible treatments.

Figure 4.

The restoration of the *Mausoleum of Theodoric* in Ravenna. A 50 mt [m?] long optical fibre delivers the laser radiation from the laser system at ground to the hand piece held by the restorer.

Figure 5. A freeze [frieze?] of the *Porta del Paradiso* by Lorenzo Ghiberti during the laser cleaning intervention. The gold film behind the dark deposit of soot, carbon particles and salts is perfectly preserved.

Laser cleaning of metals

Metals artefacts experience the action of corrosion and oxidation processes due to the many reactive agents present in the air, in the water and in the ground where these objects were kept for centuries. These are the cases of archaeological metals when they are recovered from the excavation sites, or the case of archaeological metals found under the sea. Today as well metal artefacts host in polluted environments develop typical oxidation and corrosion layers. To stabilise the state of conservation these layers have to be removed in order to apply protective coatings. Tests of laser cleaning have been reported by Pini et al. (2000) concerning Roman coins and artefacts completely covered by a thick encrustation of calcareous concretions, with oxides and salts of copper and silver. Also other bronze objects, silver objects and iron objects have been submitted to laser cleaning in many trials. Using Nd:YAG lasers in both Q-switch and microsecond mode a suitable choice of laser fluence and pulse-width could discriminate case by case the removal of the encrustations, avoiding side effects as local melting and preserving stable oxide coatings where they were present.

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An important example of the laser parameter adjustment was demonstrated for gold-coated bronze renaissance artworks, where the gilding was suffering a complex deterioration process due to environmental pollution, leading to micro blistering and loss of the gilding layer. This has been the case of the *Porta del Paradiso*, a famous masterpiece by Lorenzo Ghiberti, and the main door of the Baptistery in front of the cathedral in Florence. Many years of analysis and diagnostic studies begun in the 1980 leaded to a chemical approach based on washing with Potassium tartrate, in order to clean the outer deposits laying over the gilding layer. Unfortunately the complete washing determined a long term flourishing of salts. A laser cleaning approach could avoid the complete washing and provide an ideal cleaning if the heating of the gold film could be controlled. This case study made possible to clarify that only laser pulses ranging between 70-100 ns and a few microsecond would allow minimum transient heating to the few microns gold layer, avoiding local melting. This possibility was demonstrated by Siano & Salimbeni (2003) and now the restoration of this piece of art is going to be completed by means of laser cleaning for the last 48 freezes, one of them shown in **Fig. 5**.

Laser cleaning of paintings, paper and polychromes

For artefacts composed by materials involving organic fibres and compounds the laser approach encounters the problem of low thermal damage threshold. Furthermore the valuable historical materials have often micro dimensional structure with features of sub micron thickness, and this characteristic adds another factor of difficulty to achieve the same selectivity, precision and control well demonstrated for other inorganic materials. The situation here described is the case of paintings, paper and parchment, where the laser cleaning approach has been necessarily different from stone or metals. For the problems encountered in paintings restoration a special category of gas lasers (excimer) emitting in the ultraviolet spectrum was proposed in Greece by Fotakis (1995), to achieve sub micron removal of deteriorated varnish and also for repainting removal. In fact the ultraviolet laser radiation is absorbed in less that one micron thickness by the varnish, resulting in an adequate ablation precision.. Unfortunately most of inorganic or organic pigments react with very low thresholds to laser radiation producing an unacceptable change in the colour. In order to avoid this limit the technique developed by Scholten et al. (2000) was controlled on-line by a spectroscopic sensor (LIBS, Laser Induced Breakdown Spectroscopy) in order to detect any beginning of irradiation of the pigments layer and avoid the direct irradiation of pigments.

For the cleaning of deteriorated varnishes on paintings De Cruz at al. (2000) in USA and Italy have proposed the use of Erbium lasers at 2.9 μm, in the medium infrared spectrum. In fact molecules bearing O-H bond have an extremely high optical absorption at the Erbium laser wavelength and the radiation remains absorbed in a few microns, which is the condition for a very fine ablation control.

The application of laser techniques for the cleaning of paper and parchment has followed different solutions. For them the organic collagen structure of fibres represents the material to be preserved, besides inks and pigments constituting the graphics or the drawing. Ancient documents on paper or parchment may have problems of readability or conservation, because of accumulated dirt and dust, or fungi and other organic stains. A laser cleaning system (shown in **Fig. 6**) for high-precision cleaning of flat large area substrates has been developed by Kautek et al (1997). It allows restoration of artefacts of organic materials such as paper, parchment, leather, textiles, wood and also inorganic materials such as metals, alloys and ceramics. The laser spot is scanned over the objects through a remote computer control system. A high energy diode-pumped Q-switched Nd:YAG laser operating at 1.06 μm and 0.532 μm was installed. The authors of this system discovered that the green line at 0.532 μm was minimally absorbed by the collagen fibres, thus eliminating the occasional damage observed using the 1.06 μm line. One of the major challenges of precision cleaning was to avoid areas where ink or pigments were present. This was accomplished very precisely by image processing of the paper, verifying the ink presence and controlling the laser firing to avoid the ink irradiation.

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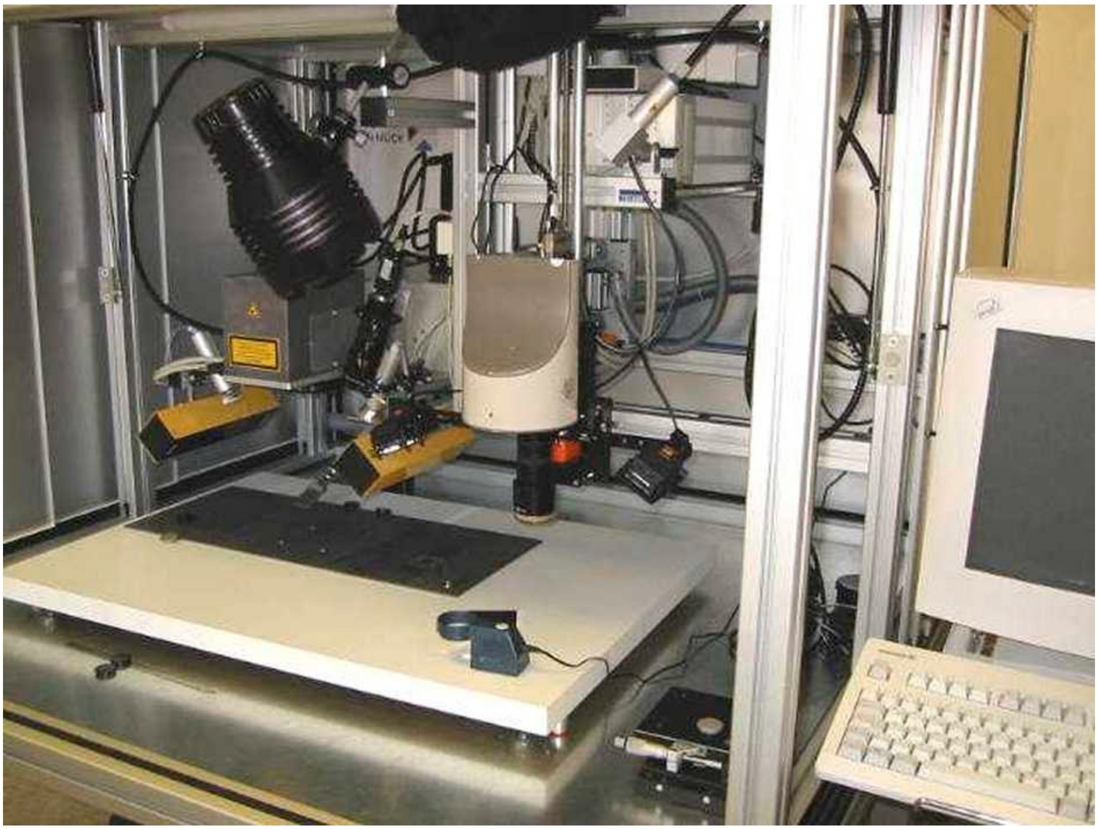

Figure 6.

The laser system developed for the cleaning of paper and parchment is organised in a closed box, providing maximum safety of operation. The set-up includes besides the laser imaging systems and a positioning table.

Conclusions

Laser techniques in conservation are presently a very interesting scientific issue, with significant successful applications and many challenges offering promising fields of research. After more than thirty years of studies and validation they are today a well accepted and appreciated professional tool in the hands of restorers. The advantages in respect with other techniques are now demonstrated, and the improvement in the precision and control of the cleaning is actually crucial especially when delicate historical layers have to be preserved. Examples are calcium oxalate on marble, gilding on bronze, paintings, fresco (wall) paintings, antique documents, antique textiles and so on. A number of well renowned masterpieces have been treated by a laser cleaning system in many countries and today the national public institutions of conservation have accepted the innovation determined by lasers in conservation. The use of the laser cleaning systems of course has to be necessarily restricted to trained restorers. The last generation of laser systems has improved the comprehension of their effects and their engineering. They allow proper settings of the emission parameters for a cleaning free of side effects, and an easy use of the hand-piece that make them to be employed as a new advanced tool.

Acknowledgements

The author wishes to thank the participants to COST Action G7 "Artworks Conservation by Laser" for the availability of their results.

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THE USE OF 3D TECHNOLOGIES TO SURVEY AND DOCUMENT ARCHAEOLOGICAL BUILDINGS AND SITE

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Kivonat

A háromdimenziós felmérés és dokumentálás terén ma a legmodernebb technológiát képviselő lézeres térszkennelés terén már Magyarországon sem beszélhetünk teljesen kísérleti korszakról. A technológia alkalmazására számos hazai példát lehet már felhozni. Az eljárás természetesen folyamatosan fejlődik, de már a mostani állapotával is az a gond, hogy többet kínál, mint amit a szakemberek be- és elfogadni képesek. Az eljárás egyértelműen a jövő hétköznapi felmérési és dokumentálási technikája, ám alkalmazásának széles körben való elterjedéséhez, az általa kínált lehetőségek komolyabb kihasználásához a felhasználói közeg gondolkodásának megváltozása szükséges. Ugyanakkor a váltás egyedi és egyszeri lehetőséget kínál, hogy egy egységes európai dokumentálási standard kerüljön megfogalmazásra és bevezetésre, amihez azonban széleskörű együttműködés szükséges.

KEYWORDS: ARCHAEOLOGY, CULTURAL HERITAGE, DOCUMENTATION, 3D, LASER TECHNOLOGY, INNOVATION, VIRTUAL REALITY

KULCSSZAVAK: RÉGÉSZET, KULTURÁLOS ÖRÖKSÉG, DOKUMENTÁCIÓ, 3D, LASER TECHNOLÓGIA, INNOVÁCIÓ, VIRTUÁLIS VALÓSÁG

Introduction

The present article is the summary of a presentation held at the conference "The use of 3D technologies to survey and document archaeological buildings and sites" in the Hungarian National Museum on 16 December 2005. Since we have expected other speakers to talk about concrete examples, we decided to analyse a general, but very basic background problem. About the technology, its methodology, the experimental results, practical application and future possibilities, see BELÉNYESY - VIRÁGOS (2004).

The laser technology doubtless represents the new generation of 3D documentation technologies. We cannot talk about experimental methods any more. Based on the surveys executed on sites – now also in Hungary – it is clear that the fundamental phase of the technical innovation arrived to its end. Concerning technology, a wide range of methods and instruments are available:

- geodesic measuring systems: total station, GPR systems
- geophysical measuring systems: soil radar, sonar, structural radar, etc.
- laser technology: 2D and 3D laser scanning
- GIS systems, etc.

Archaeology is interested in features both under and above surface, and 2D and 3D documentation systems are available for both. The most recently developed research methods are fast and provide spectacular results: there are only few specialists in heritage management, who are not convinced by the first insight. Basically, an industrial technology is transformed: the technology is developed, the field for experimental use is given, the first results provide technological feedback – everything seems to be ready for further use. However, the user side in cultural heritage management is not prepared to support the further innovation, to show direction for the development.

To sum up this introduction, the technology is given, although the development has just started. Therefore, we see the primary goal at this moment is not the separated (l'art pour l'art) technological innovation, but rather to concentrate on the side of receiver.

Some examples just to show that experimental utilisation also started in East-Central Europe (**Figs. 1-5**). Although we write about a topic with significant special literature (especially accessible on the internet), this article is a non-traditional presentation of the background problem. Therefore, the presented material is really only to illustrate what we are talking about.

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Figure 1-2.

The medieval church of Rádpuszta: side view and air view – a half-day project on site with a few days of post-processing

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Figure 3.

The medieval castle of Rezi: air view – a half-day project on site with two days of post-processing

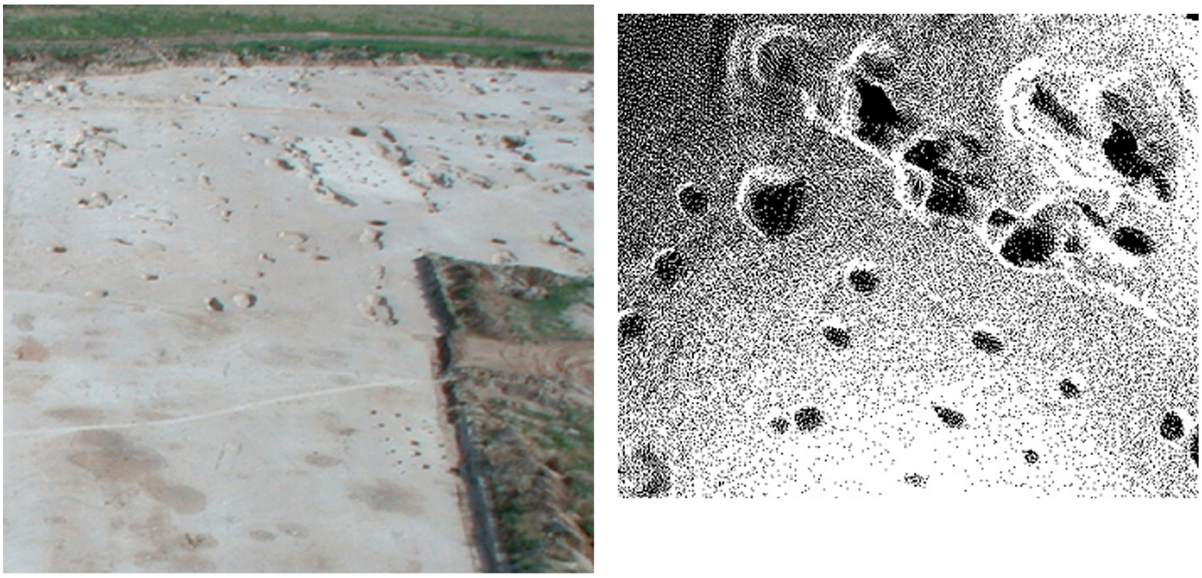
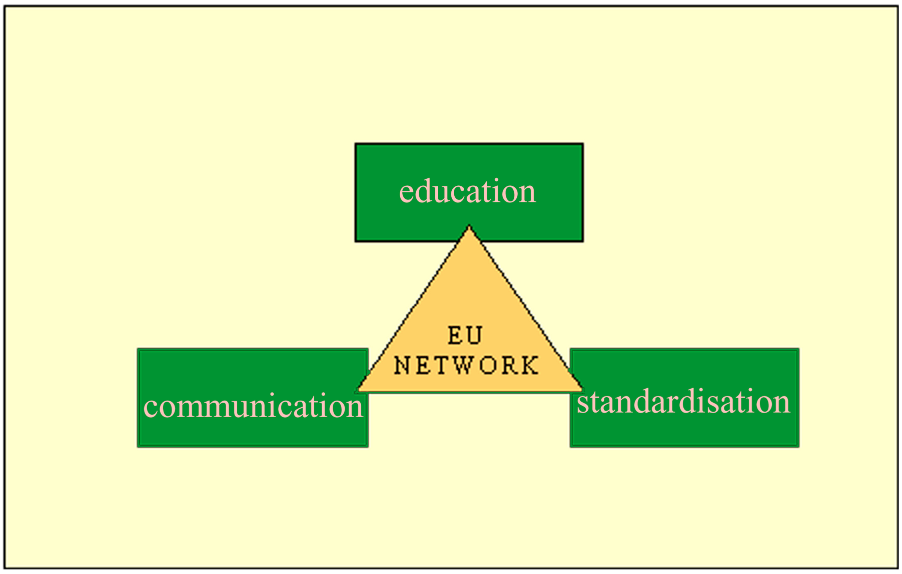


Figure 4-5.

Detail of a Neolithic settlement at Balatonszárszó: air photo and the 3D laser scanning picture (to eliminate shadow would be a very time consuming, and therefore very expensive process).

The use

E-documentation systems are now used for both documenting excavations (objects, stratigraphy, surface, etc.) and for archaeological topography (air photo, GPS, etc.). Using, however, the 3D laser technology started as an innovation. Therefore, it is looking for a market. Fast, Precise, Economical, Up to 30% quicker, Simplifies processing - the usual words in selling a product. However, from the side of the users, it would be more desirable to see the process as a new perspective of possibilities.



Figs. 6-8.

Schemes for a new model of documentation

The concept of documenting cultural heritage must be changed to fully understand the significance of the new methods. Scholars have to leave the conventional methodology to arrive to a new way of thinking. In the first case, we do our best (with drawings, photos, descriptions, etc.) to document what is there, to be able to reproduce it later.

In the other case, we can simply reproduce the reality, creating any kind of documentation later (i.e. only what is needed). Still, the expected growing in the numbers of the surveys is late. Today, both the developers and the users are looking for a market, but they often have to face uninspected difficulties.

The antipathy or aversion, which hinders the wide spread of this new documentation technology, is based on simple, but real reasons. The infrastructure of the traditional manual system is very strong, it obviously tries to withstand any of the new methods, which intend - or at least seems to intend - to change fundamentally the practise, organisation, or system of it. Therefore, accepting the new methodology is not simply a matter of fulfilling a technological prerequisite.

The major question is, whether a new attitude will emerge from the side of the heritage people to accept a new approach, a brand new way of thinking: do we want to and will we be able to apply, use, and exploit that surplus value, which is offered by the use of 3D laser technologies.

The future:

At this moment, we are facing the beginning of a revolution in the survey and documentation of standing monuments and archaeological sites. It will equally hit the technological developers and the users. To our mind it is out of question that the digital survey, data-processing, and modelling is superior to the manual versions, but the acceptance of this digital material will be decisive: what shall we do with the surplus information. This is the moment, when the users should pick up the line drawn by the technological innovation, because recently the overwhelming majority of people working on property management or in scientific research is unable to surmount this surplus information. These are just the outsiders - the so called laymen - who are obviously winning with the change, because now they are able to understand the first hand data: an easy picture to digest without almost any additional explanation.

The innovation goes on in several research centres and by several companies. Still, the possibility is given to work on a new documentation standard - and this is now or never. If the users will set up systems of documentation separate from each other, there want ever be a common, comparable, and compatible system in heritage management. The fight for an international integration of the data and data-processing requires an international network (shall we think in the EU, or wider?) and the introduction of the new technologies and the new mentality in the special education. The education and research centres should be connected in a common EU network to avoid falling into the trap that the technological development - simply because of its characteristics - was not able to escape from: to be separated and l'art pour l'art. This is not simply the objective for the future, but a real and urgent demand from both sides.

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NON-INVASIVE INSTRUMENTATION FOR DETECTION AND COLOUR CONTROL OF PAINTINGS AND ART WORKS

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Abstract

The fact that every work of art is a unique piece emphasises the necessity of working with non-invasive methodologies. In this communication instruments developed at IFAC-CNR, Florence, and their application to actual cases will be presented. Such instrumentation is based on spectroscopic techniques, namely image spectroscopy (IS) and fibre optic reflectance spectroscopy (FORS). Indeed, the combined use of these two techniques constitute a powerful tool for obtaining a large amount of spectroscopic information without any sampling, thus overcoming all the limitations and problems involved in sampling operations. Moreover, the availability of lightweight and compact equipment makes it possible to perform measurements in situ on objects that cannot be removed from their location. Furthermore, due to the fact that these techniques are non-invasive and are thus safe for works of art, it is possible to re-measure the same object after a given time, to monitor the progress of the conservation of the work of art, and also to follow the restoration processes. The methodology also enables the acquisition of a large number of spectra over the entire artefact. This wide sampling operation, the performance of which would be unimaginable with micro-sampling techniques, provides a large amount of data, which can be used for statistical analysis. As regards the application to actual cases, two case studies will be reported: a) a Leonardo da Vinci's painting, where pigments, binding medium, preparatory layer and previous restoration works were identified; b) the monitoring of the colour evolution of a Luca Signorelli's predella over the years during the exhibition to the public, during the restoration intervention and after the restoration. Finally, a brief account of the studies performed at IFAC – CNR on indoor light control will be given and the possible use of a new light dosimeter will be suggested.

KEYWORDS: IMAGE SPECTROSCOPY, FIBRE OPTIC REFLECTANCE SPECTROSCOPY, NON-INVASIVE MEASUREMENTS, COLORIMETRY, PAINTINGS

KULCSSZAVAK: KÉPEK SPEKTROSZKÓPIAI VIZSGÁLATA, SZÁLOPTIKÁS SPEKTROSZKÓPIA, RONCSOLÁSMENTES VIZSGÁLAT, SZÍNVIZSGÁLAT, FESTMÉNYEK

Introduction

The uniqueness of works of art requires analytical techniques that cause as little damage as possible to the work itself. Accordingly, two possibilities are offered to scientists for obtaining knowledge that is as complete as possible regarding an object under study: micro-invasive investigations or non-invasive investigations. Apart from possible damage caused to the object, it is quite evident that micro-invasive techniques, which can supply an accurate characterization of a single specimen, have to be limited to only very few samples, thus providing only partial information. Moreover, if monitoring of conservation state with time is required, analysis cannot be repeated exactly at the same point. It is quite evident that completely non-invasive techniques have to be preferred, even if it is honest to admit that single non-invasive techniques can be exhaustive only in particular cases. However, the combined use of several techniques is an unquestionable advantage. At present, many non-invasive techniques are available, which can give information on elemental composition and molecular or crystal structure. Moreover, many of them, besides the information on a single spot, can give 2D- or 3D-images that can be more easily managed also by non-professional people.

Here the focus will be brought on two techniques, fibre optic reflectance spectroscopy (FORS) and hyper-spectral image spectroscopy (IS). FORS is a point-by-point technique, while IS supplies 2D images. Both techniques require no sampling at all and the instruments can be transported for *in situ* measurements (museums, laboratories, restorer's atelier and so on). This aspect is very important, because many artefacts cannot be moved either for intrinsic (for instance frescoes) or for safety reasons. Moreover, the complete non-invasiveness makes it possible to repeat the measurements time to time to reveal possible alterations of the colour or of the constituting materials.

Instrumentation

As regards fibre optic reflectance spectroscopy (FORS), at present exist optical fibres that allow

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investigation of a wide range of the electromagnetic spectrum (from about 250 nm up to about 11,000 nm). Special quartz fibres can cover the UV, visible and near IR range, while chalcogenide glass fibres are suitable for the mid-IR region except the small range 2250 – 2050 cm^{-1}, where the absorption due to Se-H stretching mode can mask possible absorptions occurring in this range. Two commercial instruments, Zeiss MCS 501 and MCS 511 spectro-analysers, are used for the UV-visible and near IR regions, respectively. Different probe heads were realised in our laboratory to meet the requirements of the objects under investigation. The most used probe consists of a hemisphere, in dome of which 3 fibre optic bundles are connected with 45°/0°/45°geometry. The hemisphere is gently placed upon the surface of the object, which is illuminated by the light coming from the two bundles at 45°, while the reflected light is gathered by the bundle perpendicular to the surface. Such a device is particularly suitable for colour measurement, because specularly reflected light, which could desaturate the colour, is avoided. Usually, these probe heads are used for paintings or textiles, but they were also profitably used for 3D dimensional artefacts such as statues or archaeological objects. The mid-IR region is investigated by means of a Nicolet Protégé 460 FT-IR spectrophotometer equipped with the above mentioned chalcogenide glass fibres: in this case the geometry is approximately 0°/180°, *i.e.* the fibres, which send and receive radiation, are practically collinear. This fact can lead to a distortion of the absorption band shape, so that particular care is necessary in the interpretation of the spectra.

As regards image spectroscopy (IS), two different instruments were realised in our laboratory. From a historical point of view, the former instrument consisted of a video camera visible-near IR working in the 400 nm – 2000 nm range, in front of which a rotating wheel containing interferential filters of different width was placed. Software allowed the rotation of the wheel in a way to record the image at a given wavelength. The camera was located in front of the object, which was illuminated by two halogen lamps at 45°. By using this procedure a sequence of multi-spectral near-monochromatic images was collected and stored for successive retrieval. Then, the images could be processed using multivariate analysis techniques in order to extract most of the information they contain. Finally, the reflectance spectrum related to each pixel could be obtained. The instrumentation was built up with modular items so that it could be easily transported for *in situ* measurements. However, in spite of its advantages (transportability, friendly use) the instrument had some limits as regards sensitivity and resolution. So, a new instrument was developed in our laboratory, which is characterised by high spatial and spectral resolution, 0.1 mm and 1 nm, respectively, and by fast acquisition time (an area of about 1 m^2 requires less than 4 hours). Further details of this hyper-spectral IS scanner can be found elsewhere (Casini et al. 2005). At the present the working range is 400 nm – 900 nm, but it can be extended up to 1700 nm or even above, depending on the availability of the suitable detectors. The only drawback is its size. However studies for miniaturizing the equipment are in course.

Applications to actual cases

a) Investigation of a Leonardo da Vinci's painting

To show the potentiality of the combination of the abovementioned two techniques, here will be outlined an application to a Leonardo's painting, one of the several versions of the *Madonna of Yarn winders*. As a first approach some trials were made by FORS throughout the painting to identify the constituting materials (Bacci et al. 2005a). Information was obtained regarding pigments, preparatory layer, binding medium and previous restoration works. The experimental data reveal an oil painting with a simultaneous presence of kaolin and gypsum in the preparatory layer. Different pigments were detected such as iron oxides (probably Sienna or umber earth), vermilion and copper based green pigments (verdigris or copper resinate) by comparison with our own database of spectral data of pigments. The blue areas constituted an interesting and intriguing aspect, because, besides lapis lazuli, Thénard's blue, with its characteristic three sub-bands due to the Jahn-Teller effect, was detected in some areas (**Fig. 1**). Since this latter pigment was introduced in the artists' palette only in the XIXth century, its presence is attributed to recent restoring. Therefore, detecting where the restoration was made is for us of extreme interest. The above described IS hyper-scanner resulted to be particularly suitable for solving the problem. The images were processed by means of Principal Component Analysis, but similar results could be obtained, in this particular case, looking at the band shapes of the two pigments in the range of 450 nm – 600 nm. In fact, parabolas could fit the curves, and their radius (for each pixel) was normalized and *projected* in grey levels on the 2D image (**Fig. 2**), so that the retouched areas appeared straightforwardly evident.

b) Colour monitoring and colour control

Investigating the reflectance in the visible region allows obtaining information also about colour and its possible alterations with time (Johnston-Feller 2001).

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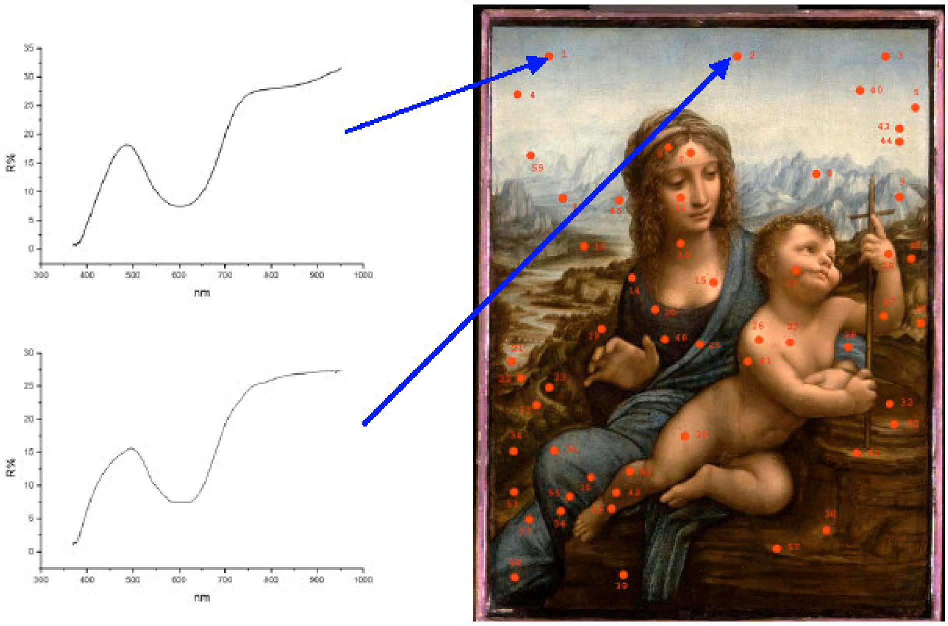

Figure 1.

The Madonna of Yarn winders. Red spots correspond to the areas investigated by FORS. The reflectance spectra of two different blue pigments, lapis lazuli (top) and Thénard's blue (bottom), are also displayed.

At this point it is worth recalling the importance of colour control both for conservation and restoration purposes. Actually, light, pollutants and other environmental factors can alter the colour of an artefact limiting the *reading* of the work or even distorting the original idea of the artist.

However, other points to be taken into account are the control of the colour during restoration works (varnish removal, colour integration, re-varnishing) and, when the restoration is terminated and the object is displayed again to the public, the monitoring of the colour evolution, which can supply useful information about environmental conditions.

Just to stress the importance of these studies for a correct conservation policy, here the experimental results of an investigation, which was done during several years, are shortly described, while a more detailed report can be found elsewhere (Bacci et al. 2005b). The work of art investigated is a *Predella* painted by Luca Signorelli in the early years of the XVIth century, which is on display in the Uffizi Gallery, Florence. The investigation cover three stages: 1) the painting was initially monitored *in situ* over a five-year period (1990 – 1995), when it was exhibited in the Leonardo's room of the Uffizi Gallery; 2) monitored during the cleaning work in the conservator atelier (2000 – 2001); 3) monitored during the new period of display to the public (2001-2004) in the museum, after the restoration and re-varnishing. The stage (1) revealed that, in spite of air conditioning and environmental control made inside the exhibition room, important colour changes occurred (Bacci et al. 1997), mainly due to the varnish layer. In stage (2) 62 points throughout the painting were investigated in February 2000 just before the restoration, in May 2000, when the old varnish was removed, and, finally, in June 2001 after the new re-varnishing with mastic resin from Chios. The latter date was the starting time ($t=t_0$) of stage (3), when a sub-set of the above points (23 points) of the painting was again examined after 18 months ($t=t_1$) and 30 months ($t=t_2$) during the new exhibition to the public. The amount of points investigated in stage (3) was reduced owing to time constraints, because the measurements could be made only when the Gallery was closed to the public. The 23 points considered were still representative of the artist's palette.

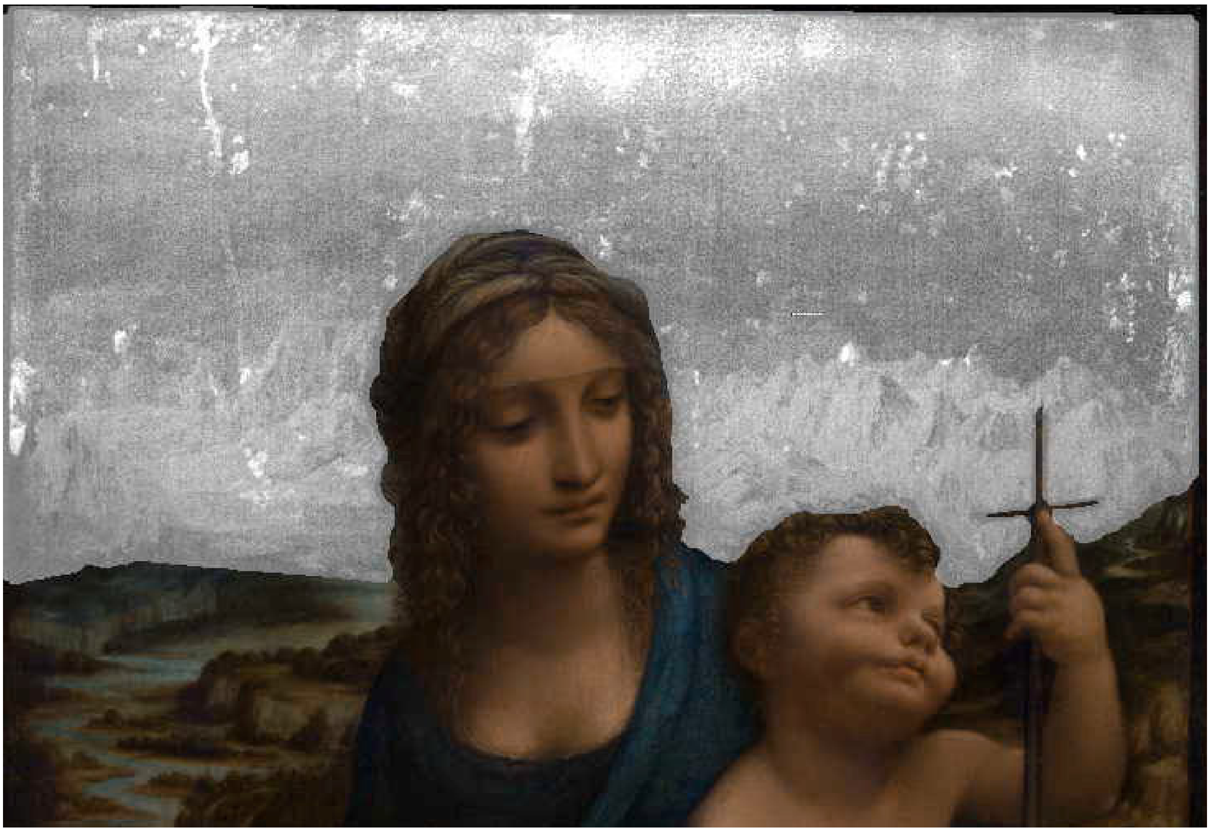

Figure 2.

Particular of the Madonna of Yarn winders. The blue areas are displayed in grey tones. Thénard's blue retouching corresponds to the whitish portions.

The removal of the varnish produced, as expected, a general desaturation of the colour and a decrease of the yellow hue, while the new varnish induced higher colour saturation (**Fig. 3**). As regards the period 2001 – 2004, a constant increase of the colour change ΔE was observed, which reached values detectable by the human eye after 30 months of exhibition to the public (Bacci et al. 2005b).

The main contribution to ΔE is given by lightness (L*), which, in general, tends to increase with time (**Fig. 4**).

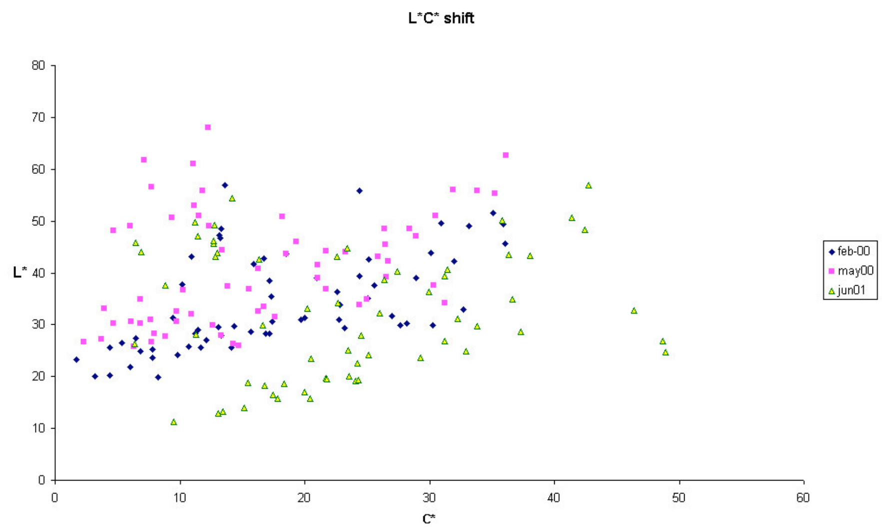

Figure 3.

The Predella by Luca Signorelli. Shift of the representative points of the areas investigated in the plane L*C*, after the varnish removal and the new re-varnishing.

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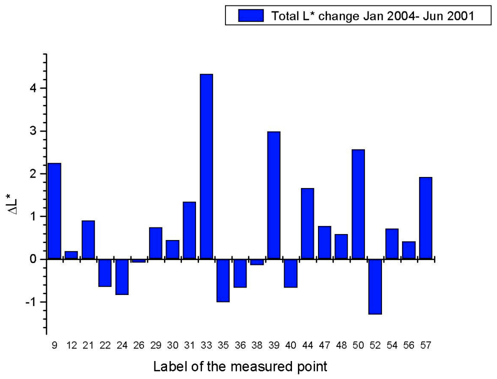

Figure 4.

The Predella by Luca Signorelli. Variation of the lightness L* for the different points that were investigated after 30 months since the new re-varnishing.

To conclude this brief survey on the scientific activity of our laboratory in the field of cultural properties, few words about monitoring lighting conditions in museums. In fact, this topic is closely related to the colour of the exhibited objects, because light is one of the most important factors in affecting colour.

The problem is: how can an efficient indoor light monitoring be arranged? What are the characteristics of an ideal light sensor? Surely, it has to be easily controlled also by non-trained personnel, it has to be aesthetically acceptable and, last, but not the least, its cost has to be moderate. To meet all these requirements, indicators were developed within a EC project under the Fifth Framework Program (Bacci et al. 2003; Bacci et al. 2005c). Small strips that change their colour when illuminated constitute these indicators. The comparison of the colour with a calibrated card, in a way quite similar to the pH indicator paper, allows a semi-quantitative estimate of the total luminous exposure.

Acknowledgement

I wish to thank all my colleagues of the Applied Spectroscopy group of IFAC-CNR together with all the students, who made important contributions to the development of the instruments and methodologies reported here.

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„FAST ARCHAEOLOGY": APPLYING NEW TECHNOLOGIES IN THE ARCHAEOLOGICAL RESEARCH IN THE CIVILIAN CITY OF AQUINCUM AND IN ITS TERRITORY

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Abstract

The sharply rising number of building investments on archaeologically protected territories in Budapest represents a particular challenge for archaeologists in the capital. The Budapest Historical Museum can only adapt itself to these demands if we apply new technologies in documentation and survey. Photogrammetry, geophysical survey and laser-scanning, to date mainly have been used to complement traditional documentation, have also been tested in excavation circumstances last year. Examples will be presented concerning the Roman villa-estates, the western and central part of the Aquincum Civil Town and its aquaduct. These types of documentation, sometimes carried out in a few hours, were not only of help to the archaeological research, but also yielded new archaeological information: that shed light on previously unexplained data from old excavations at the Civil Town of Aquincum.

KEYWORDS: AQUINCUM, URBAN ARCHAEOLOGY, PHOTOGRAMMETRY, GEOPHYSICAL SURVEY, LASER-SCANNING

KULCSSZAVAK: AQUINCUM, VÁROSI RÉGÉSZET, FOTOGRAMMETRIA, GEOFIZIKAI FELMÉRÉS, LÉZER-SZKENNELÉS

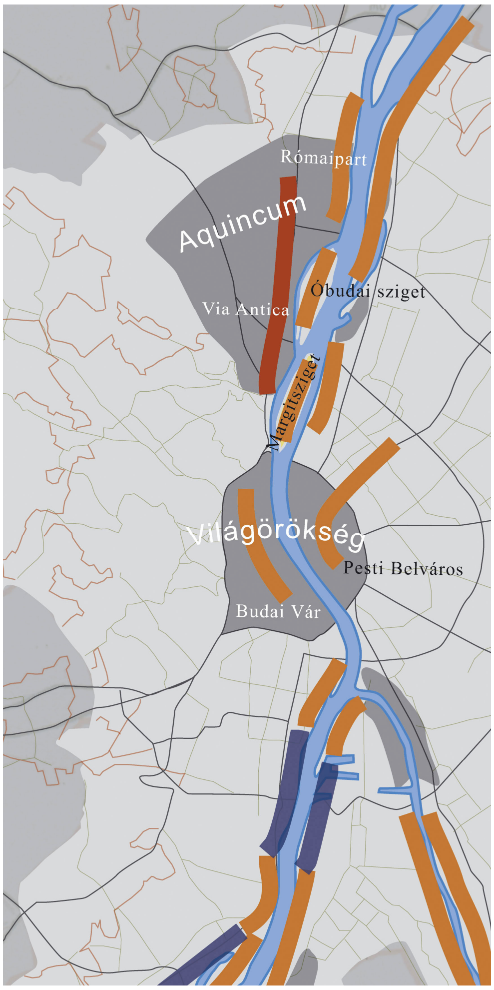

Figure 1. Historical areas in Budapest (BTM Archives)

New demands: growing number of excavations in Budapest

The growing number of building investments on archaeologically protected territories, represents an ever increasing challenge for archaeologists throughout Hungary. The situation is especially critical in Budapest, where building activities bring to light the finds and ruins of Prehistoric, Roman and Medieval Óbuda, Buda and Pest.

In the last 15 years, since the change of the regime - more than 600 excavations were carried out by the Budapest Historical Museum, responsible for the archaeological excavations all over in Budapest (Bodó 2005, 8.). As the building activity has been increasing in these years, our statistics show that the number of planned and control-excavations decreased and practically disappeared, while those connected to building activities is still steeply increasing.

Figure 2. Excavating in deep: the foundations of future buildings can destroy archaeological sites (photo of the author)

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Figure 4. Parts of the Roman settlement complex of Aquincum (BTM Aquincum Museum Archives)

These last include the building of shopping centres, housing estates or office-buildings with deep garages which once and for all destroy the cultural heritage. (**Fig. 2.**) What makes it even worse, the primary intention of investors, even if they are interested in archaeology, is to have the excavation carried out by the Museum as soon and as cheaply as possible. The demand is similar when public utilities are being constructed or in the case of public transport. Museums - and so is the Budapest Historical Museum - can only adapt themselves to these demands if they apply new technologies in documentation and survey.

New technologies: different sites- different intentions

The Aquincum Museum – in the framework of the Budapest Historical Museum – is responsible for all the Roman period sites in the capital. The capital of the province Pannonia Inferior was Aquincum, which included different settlement-units: the legionary fortress, the military town surrounding it, the Civil Town and the villa-estates up in the Buda hills. (**Fig. 3.**) As the right bank of the Danube, the Buda side, is scattered with Roman period remains, building activities also endanger the sites. To fulfil the above mentioned expectations concerning the excavations, the Aquincum Museum has long been trying to apply new technologies. Photogrammetry, geophysical survey and laser-scanning have been tried to complete traditional documentation.

Testing these new technologies took place in the territory of the Civil Town, that is to say in the area of the villa-estates and in case of the Civil Town itself as well.

Both areas required different types of technologies: the territory of the Civil Town is largely still unbuilt and undisturbed by modern building activity, so geophysical survey can be carried out. The area of the Civil Town is partly built in, mainly by modern roads and railways, so here photogrammetry and laser-scanning can be of great help, though there are areas where geophysical survey can also be helpful, as we will see below.

The territory of the Civil Town

The so- called Testvérhegy-villa

The remains of a Roman building-complex, which is located by the main northwest-southeast road, that connected Aquincum to Brigetio (modern Szőny) was first excavated in the 1930's by S. Garády and again between 2002 and 2005 connected to the building of a large housing estate. (**Fig. 4.**)

Figure 4.

Location of the so called "Testvérhegy-villa" (BTM Aquincum Museum Collection of drawings)

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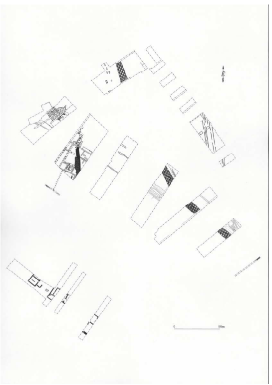
Figure 5.

View of the entrance-unit of the so called "Testvérhegy-villa" (photo of the author)

Figure 6.

Excavated parts of the villa-complex. Geodesy (BTM Aquincum Museum Collection of drawings).



During this recent campaign, parts of the above mentioned road, graves lining the road, entrance and fence of the Roman estate, building of economic purposes and parts of the dwelling-houses on the neighbouring slopes came to light (Garády 1936, 88-96., Garády 1938, 184-187., T. Láng 2003a, 95-110., 2004, 90-105., 2005a, 214-215., 2005b, 343-360, 2005c, 657-666.: **Fig. 5-6.**)

As the present owner of the site intended to reconstruct part of the Roman buildings and at the same time wanted to know how far the Roman remains extended, geophysical analyses were carried out on the slopes. The analyses resulted several "new" walls, as well as modern disturbances and also a curving structure, which can be interpreted as an apse belonging and maybe connecting two buildings (T. Láng 2005b, 357, fig. 9.: **Fig. 7.**).

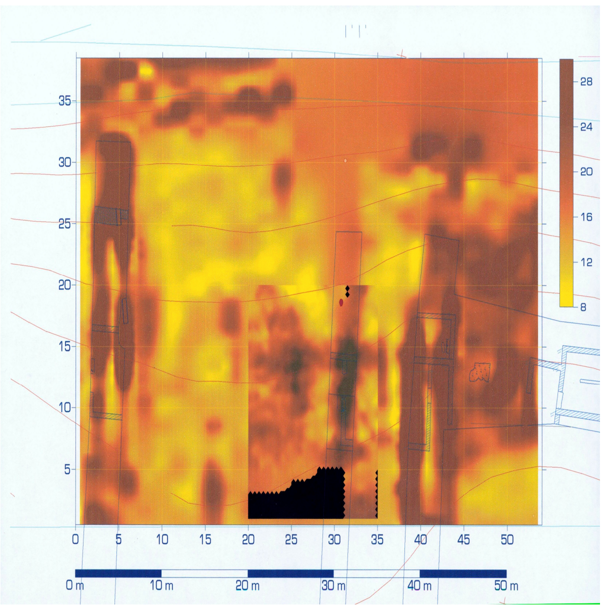


Figure 7.

Geophysical analysis of the area of the villa-buildings with the suspected apse

(Geomega Ltd.)

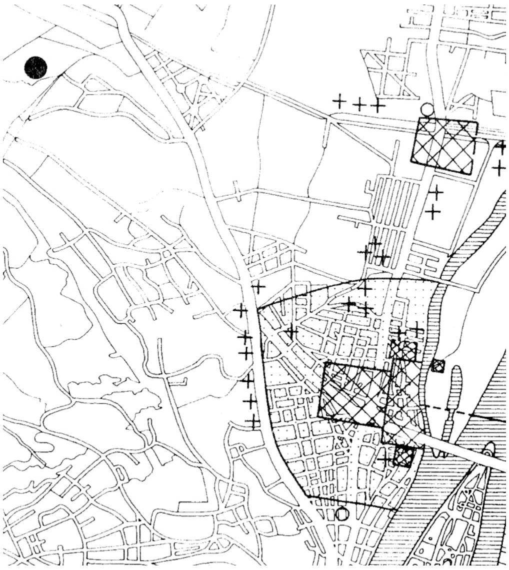


Figure 8.

Location of the “Csúcshegy-Harsánylejtő” area (BTM Aquincum Museum Collection of drawings)

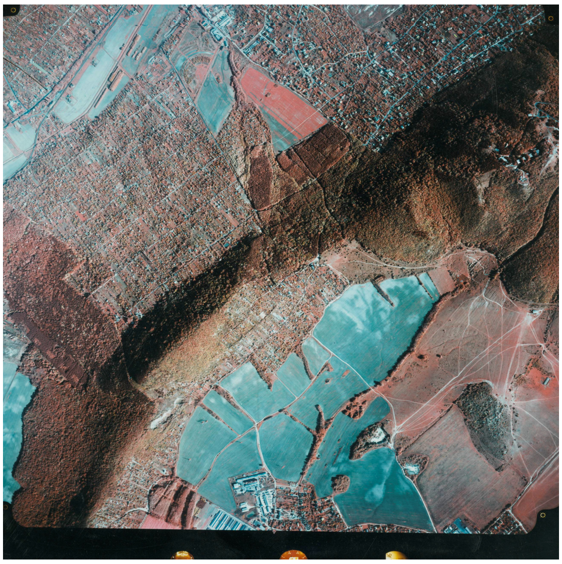


Figure 9.

Arial photo of the “Csúcshegy-Harsánylejtő” site (BTM Aquincum Museum Collection of photos)

If so, this could have been a similar main building of a Roman estate, as that of the so-called Csúcshegy villa nearby (Nagy 1937, 27-60). Excavations are still going on the slopes, thus a more complete ground plan of the building-complex could be expected (Havas 2006, in press)

Prehistoric, Roman and Medieval remains in the Csúcshegy-Harsánylejtő

North-west of the above mentioned Roman settlement-fragment, another large area was investigated by new technologies. Here, in the eastern part excavations were first carried out in 1996 by P. Zsidi, then remains of a Medieval village, part of an early Roman settlement and traces of a Roman period earth-wood construction were identified (Zsidi 1997, 58-65., **Fig. 8-9.**) Geophysical analyses were also carried out by a curving structure, close to the earth-wood construction (op.cit, fig. 23 and 24.) The analyses were carried out by Fractal Bt. (**Fig. 10.**)

Later site-surveys resulted Roman ceramics and bricks suggesting a settlement somewhere in the area (T. Láng 2005d, 216-217.)

Excavations were carried out here again in 2005 by Z. Havas, Z. Kárpáti and G. Szilas. The owner of the plot decided to build a housing estate here. Postholes and pits datable to the Bronze Age, Roman pits and remains of the above mentioned 11th c. village came to light (Havas et alii 2006, in press).

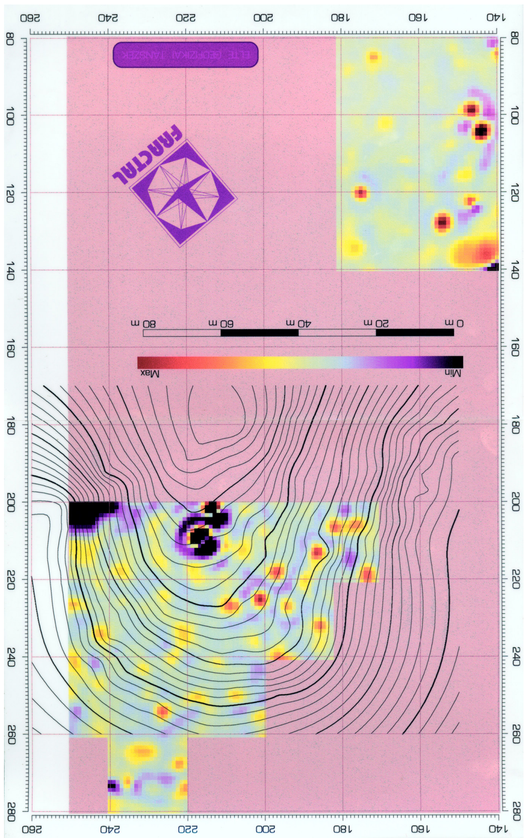


Figure 10.

Result of geophysical analysis of the earth-wood construction at “Csúcshegy-Harsánylejtő” (Fractal Bt.)

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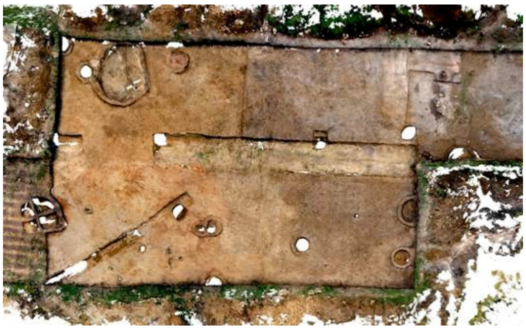

Figure 11.

Area of the 2005-year excavation at “Csúcshegy- Harsánylejtő” with Prehistoric and Medieval objects. Laser-scanned point-cloud (Piline Ltd.)

Part of the excavated area was documented by laser-scanning too, carried out by Piline Ltd. (**Fig. 11.**)

Research was also carried out in the western part of the area, where beside opening trenches, common decision was made to carry out geophysical analyses. The area is large and the results could make planning easier, avoiding the destruction of eventual Roman remains by garages or house-bases. (**Fig. 12.**). The analyses resulted possible walls and debris concentrating at the southern extremity of the site and also in the middle, which were partially identified by our trenches.

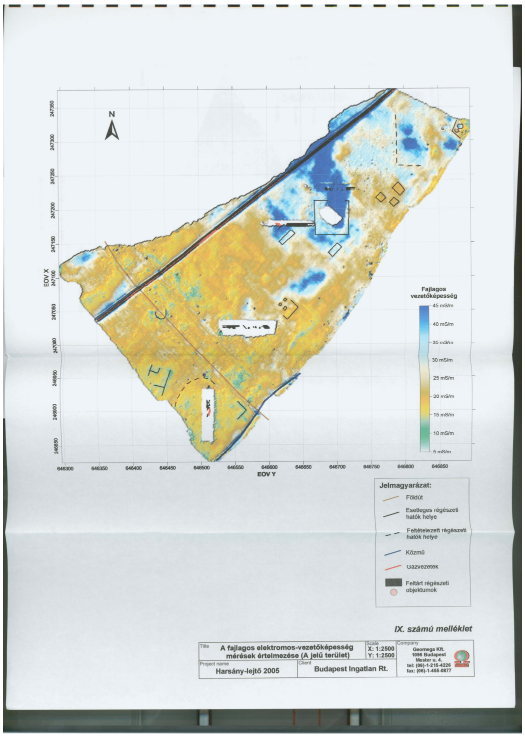

Figure 12.

Result of geophysical survey at the western part of “Csúcshegy-Harsánylejtő”(Geomega Ltd.)

Figure 13.

Walls of an unidentified building of the Roman period (photo of the author)

The analyses were carried out here by Geomega Ltd., too. Here excavations revealed Roman walls of an unknown complex, a waste-pit and a rectangular building of the same period, all in heavily ploughed condition (T. Láng 2006a, in press, **Fig. 13.**). Although the archaeological research will be continuing in 2006 as well, the geophysical data already give us an idea of the areas most intensively built in.

The Civil Town of Aquincum

Aquincum-West

The area of the Aquincum Civil Town has been continuously excavated in the past 100 years, but mostly its eastern zone.

Figure 14.

Result of the geophysical survey carried out in the south-western part of the Civil Town of Aquincum (Fractal Bt.)

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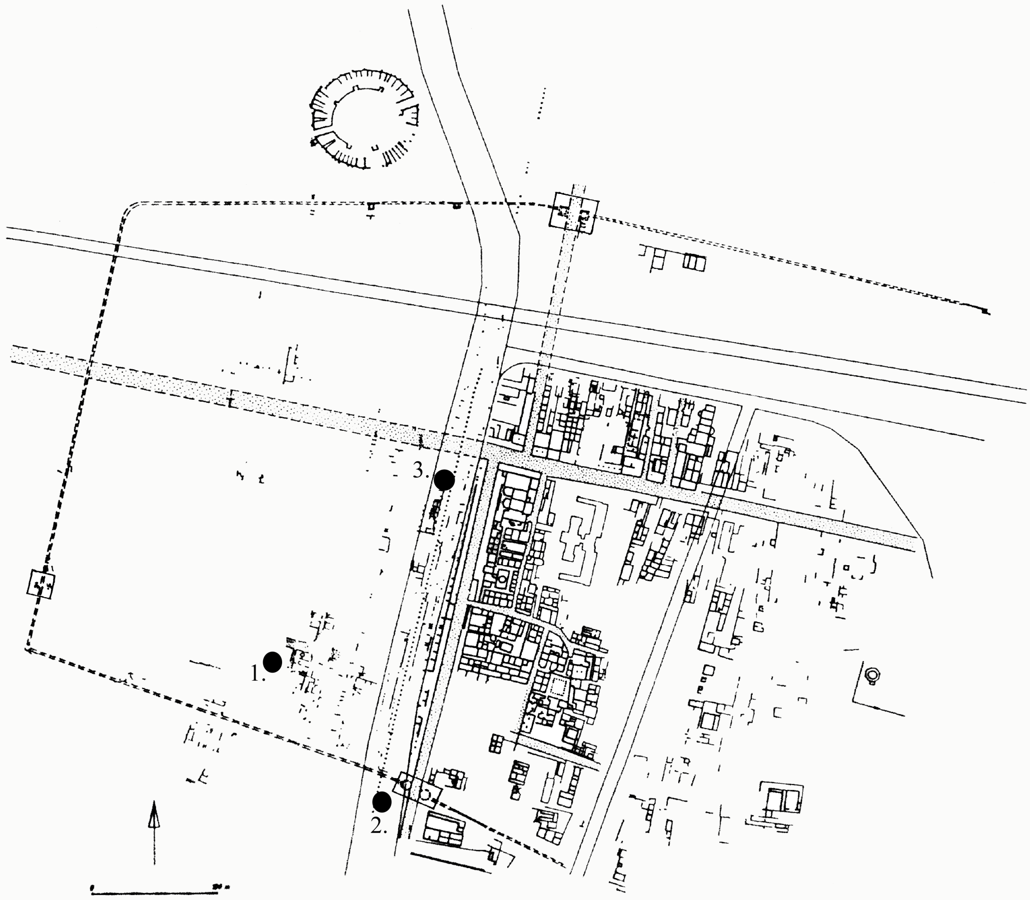

Figure 15. Groundplan of the Civil Town of Aquincum I. South-western part of the town; II. Aquaeductus; III. Railway-line

The area of the western part of the Civil Town of Aquincum is largely unexcavated, only small scale researches have been carried out here, especially in its south-eastern zone where a bath-complex was uncovered previously and by its main east-west road where shops, storerooms and workshops came to light (Póczy 1984, 21. Forschungen..2003, 151-152. and 157., **Fig. 15, 1.**) Being an undisturbed area, it seemed ideal to give a try to geophysical surveys. Although a full-scale survey is still awaiting, probes were made in the south-eastern area in the nineties. The surveys were carried out by Fractal Bt. (**Fig. 14.**) The analyses resulted a grid-system, which most probably reflect the street-system of this zone of the town. Surveys will hopefully continue in the future to give a more complete picture of the ground-plan of the western part of the town, without excavating.

The aquaeduct

The water-conduit, that supplied the Civil Town of Aquincum and the main bath- complex of the legionary fortress some 5 km to the south, originates from the area of the so called Roman Open Air Bath to the north, where well-houses were constructed and sanctuaries were built. (**Fig. 15, 2.**)

As this conduit follows the main north-south road used in all periods, the pillars of the aquaeduct were always in the focus of archaeological research. The largest systematical research was carried out between 1975 and 1979 by P. Zsidi and M. Kaba, when the Szentendrei road was widened and the pillars of the aquaeduct, its structural elements, their immediate neighbourhood together with road-fragment and shops were identified (Kaba 1976, 225-230., Zsidi 1984, 461-462.)

Figure 16.

Photogrammetrical analysis of a pillar of the Roman aquaeductus (drawing by J. Vajda and I. Győrfy)

Reconstructed pillars outside the southern town wall were “touched” again during a control-excavation in 2003 connected to a road reconstruction (Havas 2004, 61-65.). As the time was very short and only ten pillars could be studied, photogrammetry was applied to document the remains of the pillars and the limestone blocks above them.

Photogrammetrical analyses were carried out by Vajda J. and Győrfy I. (**Fig. 16-17.**) These analyses shed light on to the structure of the conduit, the statical condition of the pillars as well as to its functional problems and eventual repairs.

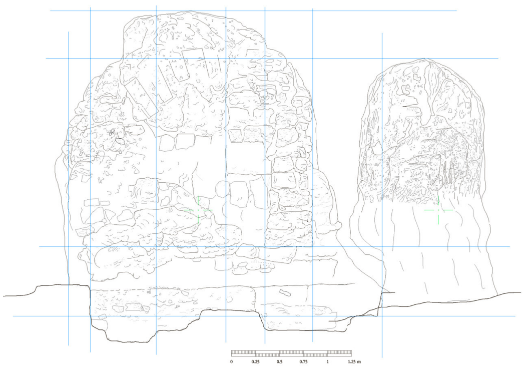

Figure 17.

Photogrammetrical analysis of a pillar of the Roman aquaeductus (drawing by J. Vajda and I. Győrfy)

Figure 19.

Result of the geophysical survey carried out during the reconstruction of the railway line in the Civil Town of Aquincum (Geomega Ltd.)

Figure 18. Excavating at night during the reconstruction of the railway line in the Civil Town of Aquincum (photo by G. Lassányi)

In spite of the fact that almost the entire row of pillars was fully excavated earlier, new data were gained from the photogrammetrical survey of the recently re-examined pillars and their immediate neighbourhood.

Centre of the Civil Town: the reconstruction of the railway-line

Applying new technologies in archaeological research in extreme conditions first took place this year. The reconstruction of the railway-line for the Szentendrei HÉV required special attention as it crosses the Civil Town of Aquincum in north-south direction. (**Fig. 15, 3**) Comparing the plans of the transport company (BKV Rt.) and information gleaned from previous excavations made it clear that the reconstruction work could affect Roman layers. The strictly scheduled project only allowed 16 hours for archaeological work to be completed The excavation was conducted by G. Lassányi, P. Hárshegyi, P. Zsidi and T. Láng. (**Fig. 18**)

Taking up the tracks meant that a circa 400 m long and 10 m wide stripe of the Roman settlement came, more-or-less, to light. The first step was a geophysical survey, which was especially important as there was no possibility to excavate deep layers. The analyses resulted walls of different orientations, roads, and intensively built in areas outside the southern town-wall. (**Fig. 19**.) Although the results of the research are still await evaluation, these are all new and essential data in understanding the topography of the town.

Following the geophysical survey, the ground surface was cleaned. At this time buildings of unknown purpose came to light together with traces of terrazzo and heating channels. (**Fig. 20.**) The documentation of this building together with other wall-fragments was carried out by laser-scanning during the night and also in rain. Beside digital and 3D images, traditional type of documentation (namely drawings) was also produced by this method. (**Fig. 21-22.**)

These types of documentation, carried out in a few hours, were not only of help to the archaeological research, but thus far unexplained data from previous years of excavation in the Civil Town of Aquincum could be explained and it was possible to further complement the topographical-chronological picture of this part of the Roman town.

Figure 20.

Remains of a Roman building with heating channel during recovering in the Civil Town of Aquincum (photo of the author)

New technologies: conditions and usability

Having tried new methods in archaeological research in Budapest, we can now outline under which conditions can these be useful. First of all it is important to emphasise, that archaeologists must see what these technologies are good for (e.g. partly replacing traditional documenting methods) and rely on the results.

As investors pressure the Museum towards fast work, it is also essential that these technologies should be quickly implemented and not only at the site, but also with the fast results.

A geophysical survey of a site or laser-scanning of a wall-structure will do not do the work alone and solve things that the archaeologist can not: that is to say the processes require continuous consultation with the archaeologist-in-charge.

There are cases when work has to be done in the rain or at night. These technologies should be used in extreme conditions: in cellars, at night, in bad weather.

Last but least: museums are known to be mostly in financial difficulties. These technologies can only be paid by the investors, as in the case of the excavations according to Hungarian heritage law. Therefore these surveys should be affordable to make owners see: it is worth using them, either because it makes documentation quicker or either because it is a faster way of getting data of a site before building even without destructive - and costly -excavation methods.

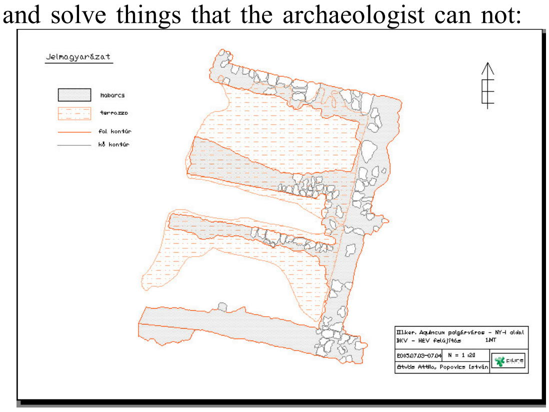

Figure 21.

3D model, based on the point-cloud of the laser-scanned Roman walls recovered during the reconstruction of the railway line in the Civil Town of Aquincum (Piline Ltd.)

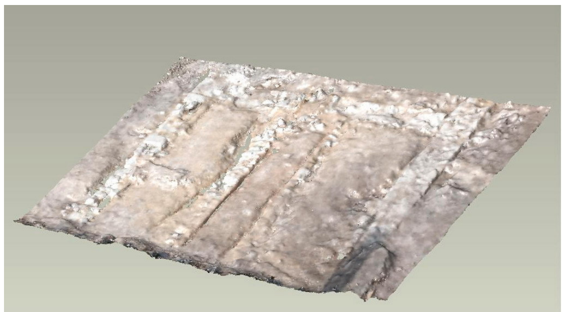

Figure 22.

Drawing based on the laser-scanned Roman walls recovered during the reconstruction of the railway line in the Civil Town of Aquincum (Piline Ltd.)

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RIVER ARCHAEOLOGY – A NEW FIELD OF RESEARCH

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KULCSSZAVAK: FOLYAMI RÉGÉSZET, MAGYARORSZÁG

The beginning of underwater archaeology dates back to the turn of the 19th-20th century Underwater archaeology became a real scientific discipline by the diffusion of autonomous diving equipment, and the work of pioneers (such as J.-Y. Cousteau, G. Bass). However these researches concentrated on marine archaeology. Even the UNESCO convention on the Underwater Cultural Heritage (2001) concentrates on marine archaeology, and only mentions the cultural heritage found in rivers and lakes.

The physical conditions of the rivers are different from that of the seas: the visibility is close to zero, the current is strong, and the depth is only a few meters. It means that the methods and technologies of marine archaeology are not applicable automatically. The role of new technologies in river archaeology is to reduce the handicap caused by the unfavourable environmental conditions.

It is possible to distinguish two groups in the technologies. The first group consists of technologies for surveying. There are a number of methods of different aims and costs. They range from the mapping of the environment to detection of buried objects. The problem of underwater documentation of finds and excavations is fundamental and needs further research.

Systematic underwater survey of rivers began in France in the 1960's, but only on a small scale. The most important among these is the work of L. Bonnamour in the Saône River (Bonnamour 2000). From the 90's the Departement des Recherches Subaquatiques et Sous-marins began the large scale surveys of the French rivers. We should highlight the Rhône, the Sein, the Saône and especially the Charente rivers (Bonnamour ed. 2000 and Dumont 2003, 2005 with further notes). The National Office of Cultural Heritage of Hungary organised an underwater archaeological research programme in 2002. We carried out surveys, geophysical prospections in the Danube, Tisza, Dráva and Rába rivers (Tóth 2002, 2004, 2005, Kérdő-Tóth 2003). In 2004 the European Fluvial Heritage programme began with Hungarian, French and Slovenian co-operation. The aim of the programme was to exchange methods, information and experiences.

The use of aerial photos and satellite technologies is spread in archaeology. In river archaeology they are useful to detect ancient river-beds, roads and bridges/ferries. In the case of the Dráva river (**Fig. 1**.) in the area of Barcs the ancient meanders, river branches are clearly visible on satellite images. Mapping these features and combination of observations with data related to underwater sites (logboats and wrecks) and archaeological maps place the sites to their paleo-environmental context.

In the case of the Dráva ship-sites it is clearly visible, that we find the underwater sites in the areas, where the modern river bed is identical, cuts or meets ancient river branches (**Fig. 2**.).

Bathymetric survey is a combination of GPS, depth measuring and cartography (Bonin 2000). The isometric mapping of the river bed could help to find shallows, artificial moles, and dams. Shallows are potential archaeological sites, because they were used to cross rivers (fords), the fast currents were used by water mills and fisheries. Bathymetric survey of the Charente river near Taillebourg (Charente-Maritime, France) found three shoals, or shallows in the environment of known archaeological finds (**Fig. 3**.). The detailed underwater survey of these areas highlighted the potentiality of these formations: a huge amount of artefacts and structures was found. Wooden posts and stone weights are evidences for fishing. A number of logboats, parts of plank-built ships, medieval weapons were also found. A further advantage of this technology is the relatively low cost and easy data-processing. The method is not able to produce high-resolution images, so it is not applicable to detect small objects, or obtain detailed images.

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Figure 1.

Satellite image of the Drava-plain near Barcs. The ancient river beds are clearly visible. (Google Earth)

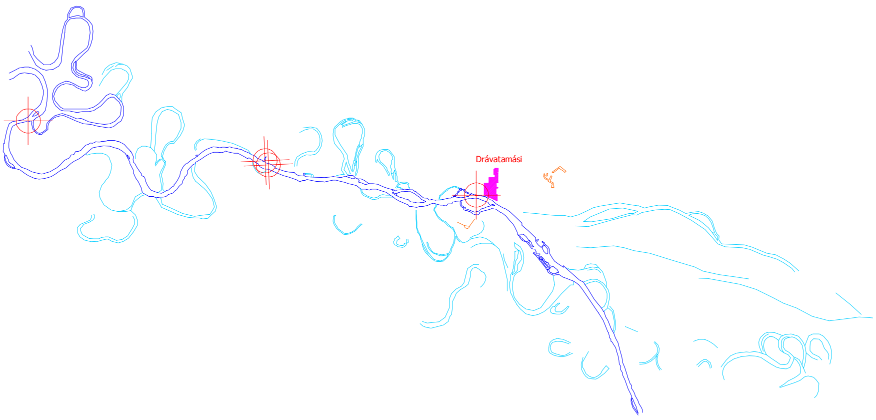

Figure 2.

AutoCAD interpretation of the satellite images in the environment of Barcs. Red points represents ship and boat finds

Sonar systems are often used in marine environment from the late '60-s (Bass 1968). Sonar (Sound Navigation And Ranging) is using the echo of sound waves, transmitted in fan-shaped beams from a moving vehicle. The echo sounds reflects the topography of the sea-bed and the computer processing of the returning waves results the image of the bottom. The system consists of a "towfish", a cable and a computer. The key characteristic of sonar is the resolution, which reflects to the frequency of the device. Marine sonar however, is not able to make images in shallow water. The new high resolution side-scan sonar (900-1200 MHz) makes good images in rivers. It makes detailed images of underwater objects (**Fig. 4**.). The problem of using this technology is that in the case of underwater vegetation-cover or intense sedimentation (tree-chunks) it is not possible to see the river bottom. Naturally the sonar is not able to make difference between a modern object and an archaeological site. Objects of small surface and low elevation (eroded wooden posts) causes problems. The sonar is a useful tool for detecting large areas during short time, but only in the case of "open" river bed. The other application area is the mapping of known composite sites (shipwreck, group of ships, walls). New, high quality fish-radar can also produce realistic images of the river-bed. We used a Humminbird 987 commercial radar in the Danube at Budapest and the Dráva at Drávatamási. At both places we could identify ships and boats (**Fig. 5**.).

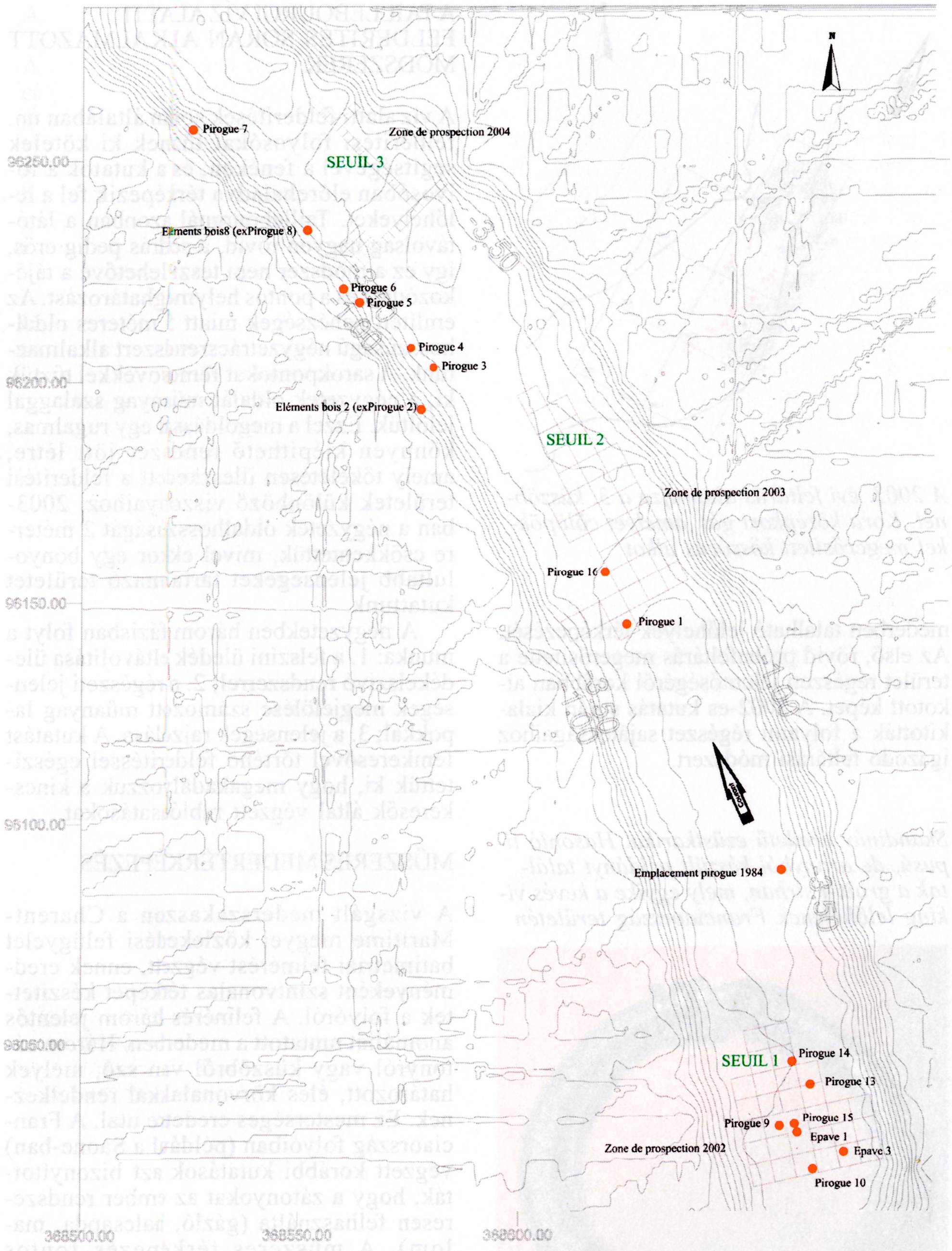

Figure 3.

Bathymetric map of the Charente river with the finds of underwater surveys, near Taillebourg (France). (courtesy A. Dumont, J.-F. Mariotti)

Seismic radar waves penetrate into the sediments. Echo sounds reflect the anomalies of sedimentation, so it is possible detecting covered structures. By the use of differentiated GPS, or constant teodolite positioning, it is possible to combine the sections made by the device (**Fig. 6.**) and create isometric or 3D images of the river-bed (**Fig. 7.**). According to our experience this is the most useful technology in instrumental survey. In the case of the Tisza at Szeged the ruins of the medieval fortification wall and the North-Eastern Tower were clearly visible on the 3D image as mounds on the river-bed. These structures stood on the right bank of the river, but they were washed over in the late 17th century. Near these structures smaller mounds were found in the river-port area, near the quay. The cross section one of them suggests artificial origin (**Fig. 8**.). Underwater survey proved, that they are shipwrecks. One of these (on fig. 7.) is a mid-19th century flat-bottomed river cargo ship, found in upside-down position.

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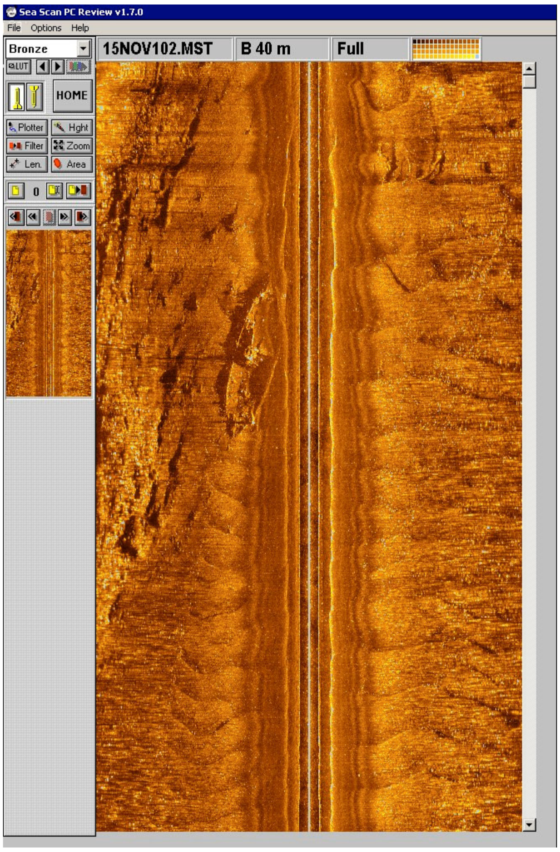


Figure 4.

Side-scan sonar image of a modern ship, Wörthersee, Austria. (courtesy C. Dworsky)

Figure 5.

Sunken boats in the Danube, near Budapest. Image produced by a Hummingbird 987 fish-radar, by L. Czakó.

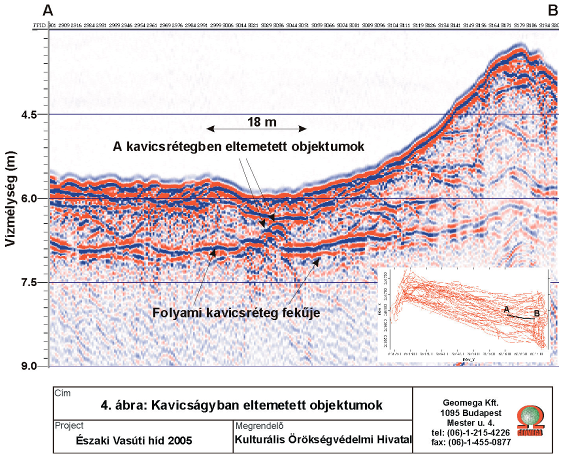


Figure 6.

A section of the Danube at Budapest, near Aquincum. An object making anomalous reflection in the sedimentation. (Geomega Ltd.)

The radar image depicts correctly the flat bottom and the curved contour of the bow/stem. This example presents that these survey technologies are useful to detect "anomalies", but the personal presence (dive) of an underwater archaeologist is necessary to identify an archaeological site.

In case of underwater survey and excavation the main problem is the orientation, the positioning of the site and the finds. The modern documentation tools of the archaeological research like GPS, laser technologies are not applicable under water. Archaeologists use hand measuring, but the scarce visibility makes it a very slow and difficult process. The SHARP (Sonic High Accuracy Ranging and Processing System) was invented in 1984 by M. Wilcox (USA). The method is based on ultrasonic technology (Shomette 1997). A grid of ultrasonic transceivers is implanted around the site, the diver emits short ultrasonic signs from a portable transceiver (used as a pen, **Fig. 9**.). The position of the "pen" is calculated from the signs detected by the fixed transceivers (all of these instruments are connected to a computer by cable). The first field tests demonstrated, that the range of the device is over 100 m, and the accuracy is 1-2 cm. This technology is used mainly in the positioning of ROV-s (Remote Operated Vehicles), but evidently useful in the documentation of underwater sites, because the accuracy and the speed is much better than by traditional hand measuring.

According to our experience a combination of sonar (or a fish-radar) and underwater survey is a fast, effective and economic solution for the mapping of underwater sites in river environment. We used these methods for the documentation of the logboat site in the Drava river (25 logboats has been found during the first campaign). In some cases, when the sediment formation is important, seismic radar is needed to detect covered structures.

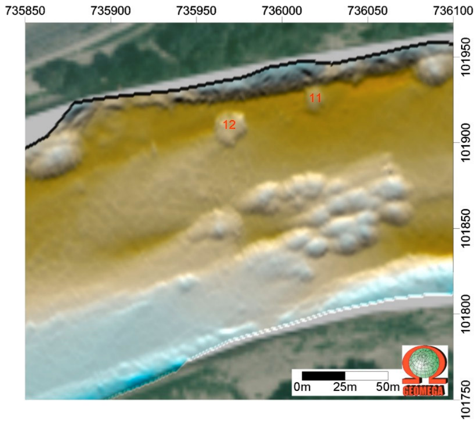
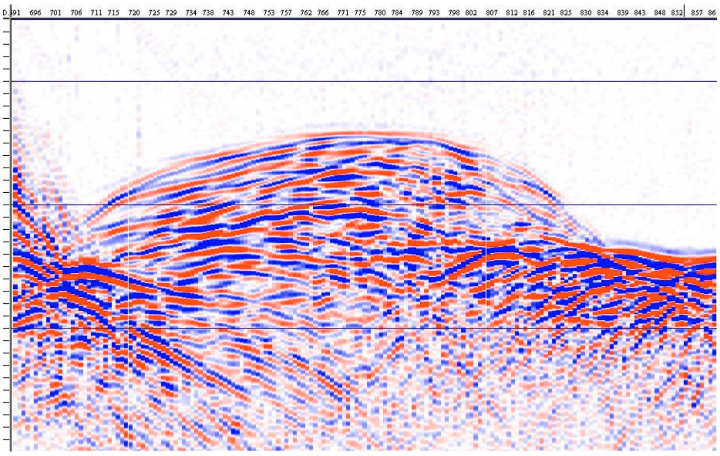


Figure 7.

D model of the Tisza river at Szeged. The mounds correspond to underwater ruins and shipwrecks. (Geomega Ltd.)



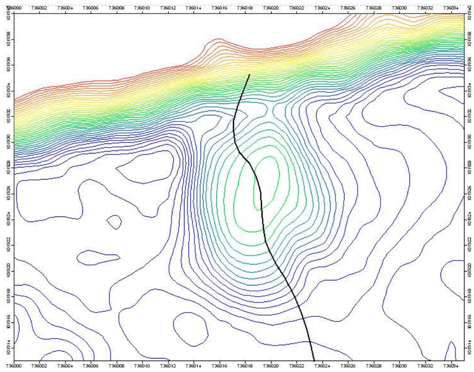


Figure 8.

Longitudial section of an anomalous mound in the Tisza at Szeged, the contour of the upside-down wreck is clearly visible. (Geomega Ltd.)

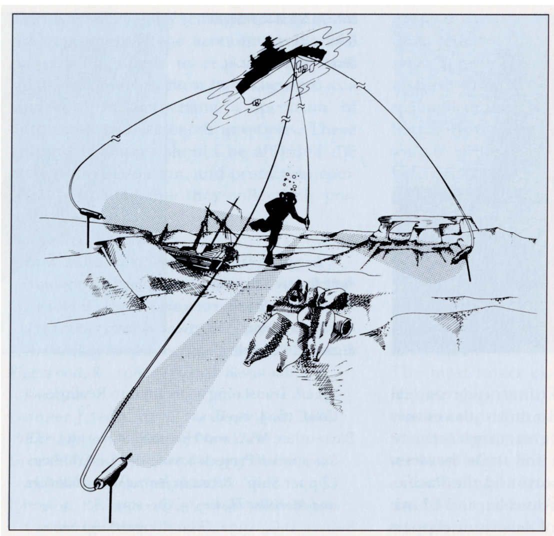


Figure 9. Sketch illustrating the working method of SHARPS system. (after Shomette 1997).

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ADVANCED TECHNOLOGIES FOCUSED ON THE EVALUATION OF THE STATE OF PRESERVATION AND RELATED RESTORATION ACTIONS IN THE MONUMENTAL HISTORICAL HERITAGE

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Abstract

*This presentation recalls a new initiative within the Emilia-Romagna Region: the establishment of the NEREA (**NE**twork for **A**dvanced **RE**newal), a consortium among research centres, universities and high-tech companies. The NEREA supports research in advanced restoration techniques and it is organised as a virtual laboratory. The main activities of the network concern evaluation of the quality of materials used in restoration and dissemination of information concerning the application techniques focused on the needs of SMEs specialised in architectural and archaeological restoration. The ultimate goal of the NEREA is creating the possibility for SMEs to certify their products at the EU level. Even though created on the regional level, the NEREA is important to the establishment of broader co-operations at both national and international level, through bilateral-multilateral agreements with parallel, specialised consortia-institutions.*

KEYWORDS: NEREA, NETWORK, ADVANCED, RENEWAL, RESTORATION, CERTIFICATION.

KULCSSZAVAK: NEREA, HÁLÓZAT, RESTAURÁLÁS, MINŐSÉGBIZTOSÍTÁS

The main architectonic features of monumental palaces, their facades in particular, while offering a direct understanding of town centres' historical development, are our very first contact with the monuments themselves. More generally they represent a cultural heritage which deserve to be preserved for the benefit of future generations. Ancient residences, however, often shows serious conservation problems, thus calling for strictly focused restoration works.

Aimed as remedy to the degradation of this unique European wealth, comprehensive analytical studies should be identified and properly implemented. The starting point is both the historical-diagnostic evaluation of the monumental building (history related research, "layers" analysis, …) and the analytical study of the physical characteristics (micro-invasive and non-destructive analyses, chemical-petrography and mineralogical surveys,…).

This will offer the necessary basis for establishing the proper intervention methodology, specific and tailored case-by-case.

Figure 1

Palazzo Bevilacqua - Bologna Photographic analysis

Figure 2.

Palazzo Bevilacqua - Bologna three - dimensional reconstruction of a constructive detail.

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Highly sophisticated techniques should be considered in the more general frame of the restoration project as a whole, because of the importance of assuring the convergence of different aspects on both the technological and cultural sides. Thus advanced diagnostic technologies are favouring the development of a new approach to the theory and the best practice of the restoration processes.

This presentation presents the recent technical development in this field and, at the same time, recalls a new initiative within the Emilia-Romagna Region: the establishment of the NEREA, a consortium among research centres, universities and high-tech companies. The centre aims at constructing two networks – one operating on the regional level and the second one on the national level – where research centres working in the area of restoration and consequent support activities can meet. The main activities of the network concern the evaluation of the quality of materials used in restoration and the dissemination of information concerning the application techniques focused on the needs of SMEs specialised in architectural and archaeological restoration. The ultimate goal of the NEREA is creating the possibility for SMEs to certify their products at the EU level. The NEREA will be committed, from one side, to the implementation of focused R&D activities according to well-proved business-related models; from the other side, to the promotion of the SMEs which operate in the specific sector. Even if created on the regional level, the NEREA is strongly interest to the establishment of broader co-operations at both national and international level, through bilateral-multilateral agreements with parallel, specialised consortia-institutions.

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