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 $5·10^7$ 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 $2.5·10^6$ n/cm^{2}s. Following the set-up of the Cold Neutron Source in 2000, we utilise a cold neutron beam of $5·10^7$ 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.

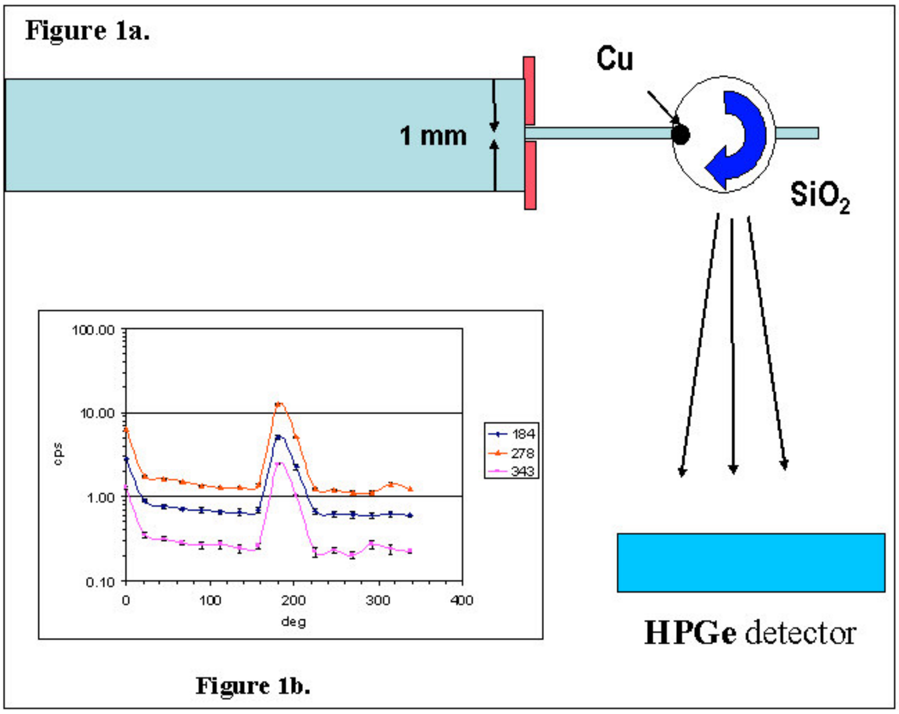

Figure 1a.

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.

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

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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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