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Neutron-based Analysis for Cultural Heritage Research.

Results of the Ancient Charm project

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Abstract: The European project Ancient Charm used non-destructive neutron-based techniques and studied a number of cultural heritage objects from Hungary, Italy and The Netherlands. The goal of the physicists in the project was to develop a 3-D imaging technique based on epithermal neutron absorption and the archaeologists wanted to use the various methods to characterize the heritage objects and, in one case, suggest methods for preservation or restoration.

Keywords: non-destructive analysis; cultural heritage; characterization; neutron study; 3-D imaging

Introduction

The study of cultural heritage objects tries to answer a number of questions like ‘what is it?’, ’how old is it?’, ‘how was it made?’, ‘where does it come from?’ and ‘how do we preserve it?’. Scientific analyses of archaeological and historic objects can help to answer these questions, and suggest methods for restoration and conservation. A large variety of chemical and physical techniques are currently employed for the characterization of objects of cultural significance. Some examples of methods are X-ray fluorescence (XRF), Particle Induced X-ray Emission (PIXE), and photon emission and absorption spectroscopy. Most conventional methods are, however, either invasive (i.e. require removal of a sample, even if a very small one) or are limited to analysis of the object surface. Such methods are widely accepted and appropriate for a whole range of materials; i.e. surface analysis will provide the best insight into the decoration of ceramic and glass vessels, and the patina and minor corrosion phases and residues on metal objects. Sampling of materials such as, for instance, production residues (e.g. slag) and building fabric is normally not a problem.

When it comes to metal artefacts however, cutting/drilling to get samples is not often permitted. Surface analyses will give an idea of the corrosion but tell us little about the composition (and microstructures) of the metal underneath. Neutrons can penetrate materials deeply and therefore thick objects can be examined in neutron-based methods. These methods are also non-destructive; it is not necessary to treat objects before measurements, to clean surfaces, or to take samples by drilling or cutting as is necessary in many other methods. This makes neutrons an ideal probe for non-destructive bulk chemical and crystallographic analysis and imaging of metal objects. The only problem one should consider with some care is the possibility that very small amounts of some elements occurring in an artifact can be activated by neutron absorption. This does not change the chemical properties, but some nuclei having absorbed neutrons can be unstable. Most of the produced radioactivity in neutron-based methods will disappear after a short waiting time. However, in cases where radioactivity with long live times can be produced, it is advisable to limit the neutron fluency to a level such that waiting time is not unacceptable long before the activity is less than internationally accepted levels below which an artifact is not considered to be a radioactive object.

Neutron methods are relevant, too, in cases where bulk analysis is needed for large intact ceramic, glass or stone artefacts, and the decision has been made not to remove samples or to disassemble the object. The constraint of carrying out work at large-scale neutron and x-ray synchrotron facilities is that the methods are non-portable: the objects must be transported to the facility.

The European project Ancient Charm used non-destructive neutron-based techniques and studied a number of cultural heritage objects from three European countries. Ancient Charm stands for Analysis by Neutron resonance Capture Imaging and other Emerging Neutron Techniques: new Cultural Heritage and Archaeological Research Methods.

Techniques

The idea of developing an imaging technique based on epithermal neutron absorption is new and presents a number of scientific and technical challenges which are best addressed by the joint development of three related 3D imaging methods: Prompt Gamma Activation Imaging combined with cold Neutron Tomography (PGAI/NT), Neutron Resonance Transmission (NRT) and Neutron Diffraction Tomography (NDT). In order to develop these new techniques the project started working with existing techniques and important cultural heritage objects from Hungary, Italy and The Netherlands. The following techniques were used: Prompt Gamma Activation Analysis (PGAA), Neutron Tomography (NT), Time-Of-Flight Neutron Diffraction (TOF-ND) and Neutron Resonance Capture Analysis (NRCA).

PGAA and PGAI

Neutrons in the cold energy range (namely of the order of a fraction of eV) can be captured by various isotopes in the sample. Prompt Gamma Activation Analysis (PGAA) (Molnar 2004) is based on the fact that, after neutron capture, many nuclei de-excite with a prompt emission of gamma radiation of characteristic energy. The gamma spectrum can be measured by high-resolution gamma-ray spectrometers, and the intensities of the emission peaks provide quantitative identification of the emitting nuclei (Figure 1). The information about the nature and concentration of elements in the sample is thus obtained from a gamma-ray energy spectrum. PGAA is a completely non-destructive technique, allowing for studies on cultural heritage objects without the need to take samples or to remove patina from a metallic surface. For PGAA analysis, the intact object is placed in the neutron beam. In the past, the number of PGAA applications have been limited by the need of a dedicated station at a neutron source like for instance the PGAA facility at the Isotope Institute of the Hungarian Academy of Sciences (HAS-IKI) in Budapest (Hungary) (Kis *et al*. 2008) or the PGAA station at the Forschungsneutronenquelle Heinz Maier-Leibnitz (FRM-II) in Garching near Munich (Germany) (Kudejova *et al.* 2008). In order to have precise results, PGAA requires the use of high-resolution detectors like high-purity Ge detectors, which in turn have the disadvantage of limited detection efficiencies and are prone to radiation damage by high neutron fluxes.

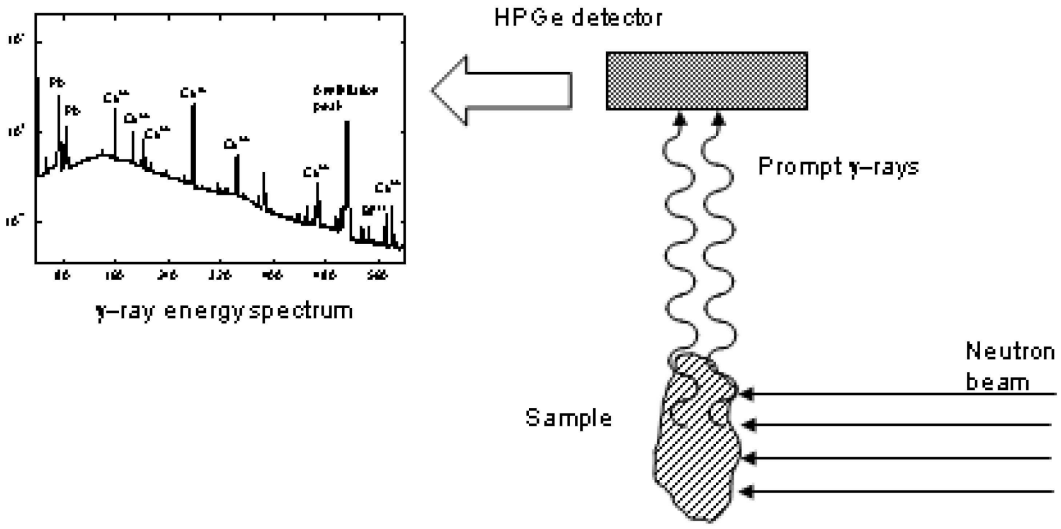

Fig. 1 – The PGAA working principle (see text for details).

PGAA can be considered as a bulk method, i.e. it measures the average elemental and isotopic concentrations in the irradiated volume; such a volume is often in the order of few cm^3. In order to obtain space-resolved information, it is necessary to scan the sample with a collimated neutron beam *and* gamma-ray detector: this method is called Prompt Gamma Activation Imaging (PGAI). In PGAI (Figure 2) the information on elemental and isotopic composition is specific of every isovolume, i.e. the region of the crossing of the neutron and gamma-ray collimators lines-of-sights (Kis *et al.* 2008). This region can be as small as few mm^3, this value giving the limit of the available spatial resolution of the technique. Compared to PGAA, the imaging method PGAI requires longer measurements, due both to the lower neutron and gamma fluxes of the collimated beam; typical values for acquisition times are 200 – 3600 seconds per spectrum (Kis *et al.* 2008). Because neutron capture cross sections for most metals are usually higher in the cold and thermal region, PGAA and PGAI are well suited for reactor-based neutron sources, which have maximum neutron flux in this region.

In order to optimise the possibilities offered by PGAI, the best approach is to combine it with neutron tomography. Neutron Radiography (NR) and Neutron Tomography (NT) are well-established techniques, whose importance for archaeology is widely known since many years (Schillinger *et al.* 1996). NT allows the 3D imaging of the interior of bulky objects made in a large variety of materials, rapidly reconstructing the shape and dimensions of internal structures with high spatial resolution (much less than 1 mm^2) and in relatively short times (typically from few minutes to few hours for a complete scan). However, it has no elemental sensitivity; in other words, NT cannot be used to determine the composition of specific artefacts in the investigated samples. By using the NT image of a sample, one can select the most interesting regions to be studied with PGAI. Moreover, the well-resolved images obtained with NT are a useful guide for the scanning and positioning of the object with the collimated neutron beam. This combination of the NT with PGAI speeds up considerably the effective scanning of the object (Belgya *et al.* 2008).

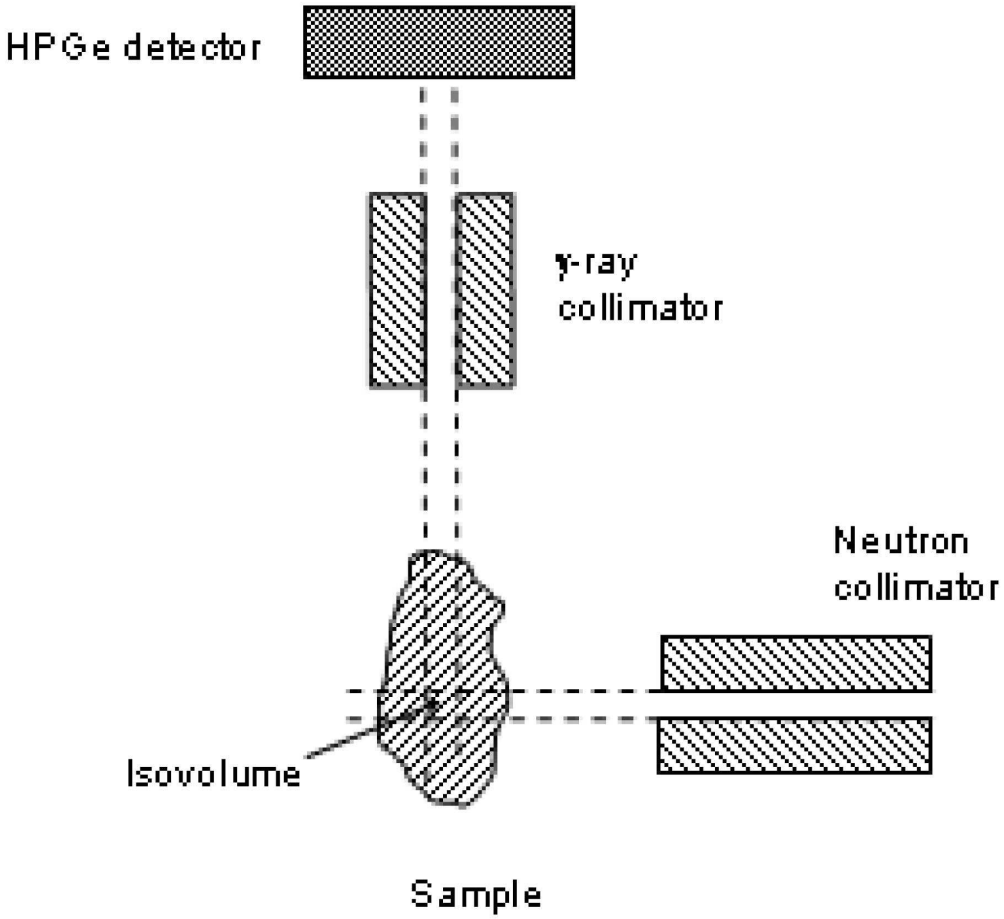

Fig. 2 – In PGAI the space resolution is given by the use of collimated neutron and gamma-ray beams. The crossing of the lines-of-sights of the collimators gives the isovolume, i.e. the space region inside the sample to which the compositional information is related (see text for details).

NRCA and NRT

Neutron resonance capture analysis (NRCA) and neutron resonance transmission (NRT) are relatively new methods based on neutron resonant absorption. The neutron cross section of many elements show intense and narrow absorption peaks in the epithermal region (E_n up to 1 keV). These peaks (resonances) are specific to every isotope and constitute as a useful “fingerprint” of the sample composition. NRCA and NRT make use of the time-of-flight (TOF) technique applicable at facilities with pulsed neutron sources to measure the resonances energy. In the epithermal regime the neutrons are non-relativistic and their energy can be expressed by the classical relation

$$E_n = \tfrac{1}{2}\, m_n\, (L/t)^2$$

(Formula 1)

where m_n = 1.675 x 10^{-27} kg is the neutron rest mass, L is the neutron flight path from the source to the sample and t the time to cover it (time-of-flight). The TOF technique is in general better suited to spallation

neutron sources like the ISIS pulsed neutron and muon source at the Rutherford Appleton Laboratory in Chilton (UK). Another option is the GELINA facility in Geel (Belgium), where neutrons are generated in pulses with a short time uncertainty. Here the neutrons are produced by stopping high energy electrons in a target of uranium. GELINA has a better resolution then ISIS; ISIS has a considerably larger flux. Opposite to PGAA, TOF-based techniques do not produce energy but time spectra (Figure 3), and they do not need detectors with high energy resolutions, but instead high time resolution. For instance, the ANCIENT CHARM instrumentation is based on Li-glass neutron detectors (Schooneveld *et al*. 2009) and Yttrium-Aluminium Perowskite (YAP) gamma-ray detectors (Perelli Cippo *et al*., in prep.), whose characteristics are high detection efficiency, low sensitivity for neutrons and neutron radiation hardness.

ANCIENT CHARM conjugates the two approaches to the neutron resonances analysis: the well-established NRCA (Postma *et al*. 2001) and the NRT.

In NRT the neutron beam transmitted through the sample is recorded as a TOF spectrum and the resonances appear as characteristic dips, whose energy position can be identified via relation (1). After neutron capture, the nuclei, left in an excited state, de-excite with the emission of prompt gamma rays that can be revealed by suitable detectors. In NRCA, however, the energy of the emitted gamma rays is not the relevant parameter; only the arrival time of the gamma photons on the detectors is tagged and recorded in a TOF spectrum (figure 3). Resonances appear in the NRCA spectrum as characteristic peaks, uniquely identifying the emitting nuclei. NRCA and NRT give thus complementary measurements of the same quantity, partially compensating their respective advantages and disadvantages. NRCA is more sensitive to many elements, while NRT is a space resolved technique, allowing for 2D images of the sample. In fact, the use of a Position Sensitive Neutron Detector specially developed for ANCIENT CHARM (Schooneveld *et al*. 2009) allows for 25 mm x 25 mm element-sensitive radiographies of the sample, with a 2.5 mm resolution; a scan of the sample can be performed in order to obtain a 3D image of the whole investigated object with standard tomographic reconstruction techniques. Typical times for a complete NRT scan of objects of the dimensions of a fibula are of the order of 24 hours.

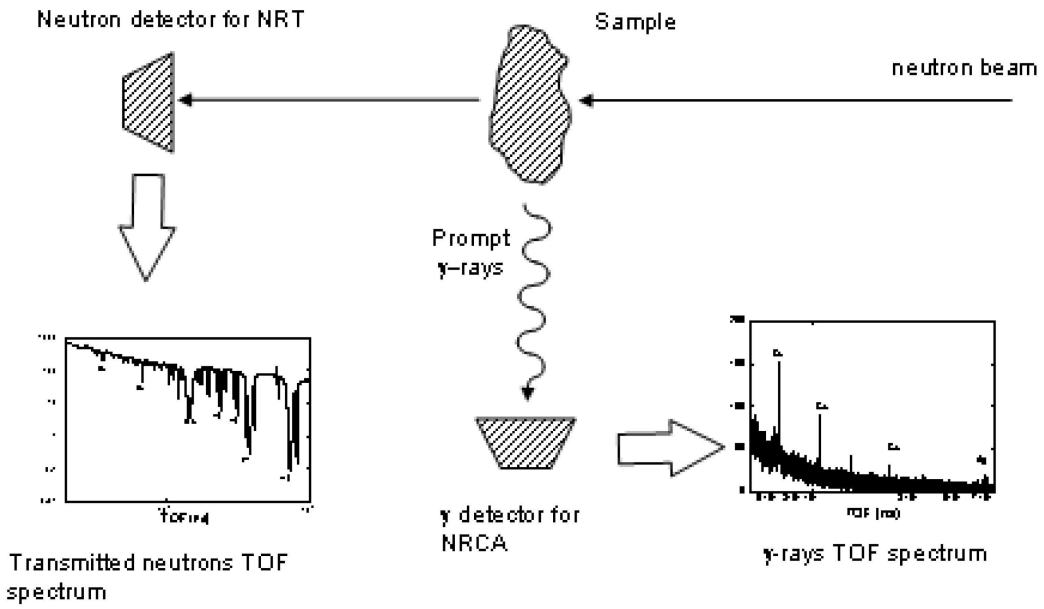

Fig. 3 – NRCA and NRT working principles (see text for details).

ND and NDT

Neutron Diffraction is based on the property of thermal neutrons to be scattered by structured arrangement of atoms, like for instance crystalline planes inside the sample, following the well-known Bragg's law:

$$\lambda = 2\, d \sin\theta$$

(Formula 2)

where λ is the wavelength of the neutron scattered at an angle θ and d is the distance between adjacent crystal planes. The neutron wavelength (in Angström) is related to the velocity v (in km/s) by

$$\lambda = 3.956/v$$

(Formula 3)

The velocity of the scattered neutrons can be easily measured with the TOF technique for every scattering angles. The diffraction process is strongly related not only to the nuclear properties of the sample, but also on the distances and relative positions of the atoms, their long-range ordered or non-periodic structures. ND gives thus information not on the nature of single nuclei, but on collective properties like for instance the crystalline or amorphous (glass-like) nature of the sample, its texture and microstructure (Kockelmann *et al*. 2001), the presence of residual stresses in metallic objects (Santisteban *et al*. 2002). These properties can be often related to the deformation processes and to the treatment history of the material (Siano *et al*. 2006): it is thus a complementary technique to the previously exposed ones.

TOF-ND makes use of thermal neutrons, and it is a well-suited technique both for reactor-based sources and for pulsed sources. Typical times to achieve satisfactory data statistics strongly depend on the sample

dimensions and neutron spot size and intensity, ranging from a few minutes to several hours. Neutron diffraction experiments are typically performed on several analysis points, in order to check for homogeneity of the material or to survey a composed sample.

In order to extend the TOF-ND to an imaging technique (Kockelmann and Kirfel, 2006), two different approaches can be used: 1) point-by-point scanning with a collimated neutron beam or 2) 2D-resolved Bragg-edge transmission. Both methods have shown their potential in metallurgical and engineering applications (Santisteban et al. 2003). In the first approach, the use of neutron collimators for both the incident and scattered beams allows to concentrate the diffraction analysis on an isovolume as small as 0.5 mm x 0.5 mm x 0.5 mm. Specialised diffractometers (“strain-scanners”) like the ENGIN-X beamline at ISIS offer the structure and phase mapping as a standard procedure: diffraction scans can be performed along arbitrary trajectories, for example into the bulk of the sample or along its surface, in order to map the strain distribution, the phase content, or the texture. This procedure, however, is quite time-consuming, and the runs are usually performed on selected parts rather than on the full sample. Crystallographic phases can also be mapped on a conventional TOF diffractometer by scanning the sample in front of a collimated beam along three directions x, y, z, using a laterally collimated beam and diffraction signals to reconstruct the positions of phases. In this case, there is no secondary collimator to confine the diffracting volume along the beam direction, and the diffraction intensity from the whole illuminated path (chord) is recorded. In order to achieve a complete information on phase distribution, the scan has to be repeated after 90° rotation.

In the second approach, phase analysis is performed in a radiography type set-up, i. e. with a position-sensitive detector in a transmission geometry. Because the neutron removal operated by Bragg scattering in the sample, the transmission detector measures a TOF spectrum which contains drastic steps in the transmitted intensity distribution (Bragg Edges, BE). BE appear when, for a given family of crystal planes, the Bragg angle 2θ equals 180°. BE can be indexed and analysed like d iffraction patterns (Santisteban et al. 2003). Use of a position-sensitive detector allows obtaining, as in case of NRT, 2D-resolved images of the phases in the sample, with the same typical resolutions.

Archaeological results

The results of research to characterize objects can help to answer questions about an artefact's historical and cultural importance. These questions can be about provenance, technology, authenticity, function - functional or ceremonial -, etc. The results can also suggest methods for preservation or restoration. The Ancient Charm collaboration selected a number of objects suitable for the development of the new imaging technique but also with research questions belonging to the list above. Six objects from Italy, Hungary and The Netherlands were chosen, two from each country.

The Gates of Paradise

The example from Italy is a non-destructive integrated neutron study of two Lorenzo Ghiberti’s reliefs from the doors of the Florence Baptistery. The study was executed to solve conservation problems. The ‘Battistero di S. Giovanni’ (Baptistery of St. John) in Florence (Tuscany, Italy) is a religious building which has the status of a minor basilica. It is renowned for its three sets of artistically important bronze doors with

relief sculptures by Lorenzo Ghiberti dating from the 15th century. Michelangelo named these doors "the Gates of Paradise" because of their beauty, and they were said to hallmarks' the beginning of the Renaissance.

The advanced deterioration state of the mercury-amalgam gilded sculptures suggested specific strategies for urgent restoration, especially addressing suitable cleaning treatments. The gilding is covered by encrustations due to a mixture of corrosion products and deposits (gypsum, quartz, feldspars, carbon, etc.) as a result of the interaction with the outdoor environment (Fiorentino *et al.* 1982, Mello 1986, Mello *et al.* 1982, Siano *et al*. 2006). As a consequence oxide and salt layers constitute the current 'bed' of the gold layer, requiring special attention from a restoration point of view. Copper corrosion compounds (i.e. copper oxides, chlorides and sulphates) are located immediately below the gold layer. Restoration over the last 20 years has focused on selectively removing some of the dangerous corrosion compounds under the gold layer and the black crusts above it. In the case of the east door two types of cleaning methods were chosen: a chemical agent bath and laser ablation.

Two objects were chosen for research: the *prophet head* from the paradise (east) door and the *north head* from the north door. The *prophet head* dates from between 1425 and 1452, the material is gilded bronze, the diameter is 13 cm, the height is 7cm and the weight is 2700 g (figure 4). The *north head* dates from 1412, is also made from gilded bronze, the size is 12 by 9 by 9 cm, and the weight is 1800 g (figure 5).

Fig. 4 – The *prophet head* from the east door of the Baptistery of St. John in Florence (Tuscany, Italy).

Fig. 5 – The *north head* from the north door of the Baptistery of St. John in Florence (Tuscany, Italy).

The aims of the study were to establish the state of the gilding and its distribution, to study the copper corrosion, to study re-melting extension inside the *prophet head* (and, eventually, the presence of other

materials inside) and, again for the *prophet head* only, to study the differences between the primary and secondary melting in terms of composition and working processes.
The surface and bulk of the artefacts were thoroughly investigated using several non-destructive neutron techniques. Prompt gamma activation imaging (PGAI) was used to derive the distribution of elemental analysis and high-resolution neutron radiography (NR) for the study of internal features. Neutron diffraction and strain measurement experiments allowed deriving the bulk phase composition of the alloys and the working methods. Finally neutron resonance capture analysis combined with neutron resonance transmission (NRCA/NRT) to identify the elements in the object and provide their spatial distribution.

For the PGAI/NR analysis four positions on the *north head* were chosen covering the three cleaning-areas. Each position was measured at different depths in 0.25 mm steps starting from the surface. Chlorine was not detected in the areas treated with chemical agents but was detected in areas treated with laser ablation. Copper chlorine proved to be dangerous for the gilding since it forms excrescences (Festa *et al.* 2009).
For the *prophet head* neutron radiography provided the extension of the re-melted area. The results suggest that, because the original melting was not satisfactory, the sculptor made a second one. The pouring of metal inside the original cavity from the rear side accumulated the casting in the front part of the head, leaving part of the original cavity empty in correspondence with the nape. The radiography revealed the presence of a light zone that was ascribed to either a hole or a fusion clay. The former hypothesis was confirmed by further experiments where a set of neutron diffraction scans performed on the artefact revealed a 3D image of the hole extension. (Festa *et al.* In prep.).

A neutron diffraction investigation was carried out at ISIS on both melting volumes. Variations in diffraction peak positions and shapes reveal a modification of the working method. Most probably, the original alloy was significantly more heat-treated than the re-melted part. The re-melted part shows evidences of a much higher cooling rate and for this reason it did not have enough time to homogenize (Festa *et al.* In prep.).

Migration period (Early Medieval) grave goods from Hungary

Two objects were selected from the collection of the Hungarian National Museum. Both objects are composed of various materials and the main research question was to determine their elemental composition and production technology. For both objects NT and PGAA bulk analysis were done at HAS-IKI in Budapest, NT and PGAI at the FRM-II in Munich (Germany), and NRT, NRT and ND at the ISIS pulsed neutron and muon source at the Rutherford Appleton Laboratory in Oxfordshire (UK). The results from Budapest and some preliminary NT results from Munich are included.

The first object is a medieval disc fibula from the cemetery Kölked-Feketekapu (Kiss 1996)(figure 6)**.** It has almandine inlays and an iron band around it. This type of fibula was the characteristic jewellery of the 6th century AD. The type presumably originates from the workshops along the Rhine in the territory of the Franks. The round ground shape with radial divisions can be dated to the middle 3rd of the 6th c. AD. Filling the middle round cell with a pearl or imitation with calcite or glass bead is a common phenomenon. The disc fibulae with garnet inlays are common in Frankish cemeteries around the lower Rhine, but do also occur in

Alamannic cemeteries in Southern Germany. They even occur in Longobardic cemeteries in the Carpathian basin showing the excellent western connections of the Longobards. The fibulae were found in exactly the same place where they are usually found in Frankish graves. It shows that also in this area they were used to close the women's shirt on the neck and that they were usually worn in pairs. The questions for this particular object were:

- Are the cells made of silver?
- What is the material of the filling of the cells?
- What is the material of the white pearl?
- Is the iron band reparation?
- Is it the product of a Merovingian workshop?

According to the first bulk material analysis the cells of the fibula are made out of gold which is very rare among this type of fibulas, about 95% were made out of silver. After the NT analysis at FRM-2 it is clear that the fibula's strange structure is due to ancient reparation. There are 2 layers of filling materials that can be seen below the cells: one ends in the lower line of the cells: the original filling paste. Below this another can be seen that fills the whole area of the iron ring: so we can preliminary conclude that when the former back plate was detached by accident and that the addition of an iron band around the fibula was to prevent the cells from falling apart. Then the remaining space was filled with another paste, and finally a new back plate with a new needle-case was attached to the fibula.

Fig. 6 – A medieval disc fibula from the cemetery Kölked-Feketekapu (Hungary).

The second object is a belt mount with metal and glass inlay (figure 7). This belt mount belongs to a Merovingian type of belt set coming from the Környe cemetery in the Carpathian basin and dates from the first third of the 7th century AD (Salamon and Erdélyi 1971). It is a triangular iron belt mount with silver messing and glass inlays decorated with a Merovingian type geometrical motif. The parallels of this type of belt were commonly used all over Western and South Western Europe in the second quarter/middle third of

the 7th century AD. In the Carpathian basin exactly the same shapes of mounts were used, only a strap end added due to the local influence. The metal inlays were made mostly according to the producing technology of the western mounts: spiral wires were used and hammered into mostly punched niches without any fixing material for the line patterns. For the plaque patterns another method was used: in Western Europe similar spiral wire was used to fill bigger surfaces, but in the example from Környe silver plates were cut into the exact form and then hammered. For the brass inlays again spiral wires were used in Western Europe, but for the Környe belt a copper cell was made for the brass. Glass inlays on iron dress accessories are rare in this period: they are frequent along the river Rhine, and some pieces occur in the Carpathian basin with other objects related with the archaeological material of the lower Rhine territory. The motives of the metal inlay, the so-called *Leiterbandtauschierung* were widely in use in the Merovingian world, but some small details of the Környe belt show a possible local production.

Fig. 7 – A medieval belt mount with metal and glass inlay from the Környe cemetery (Hungary).

The research questions on this object were mainly about characterization and production: how were the brass plates fixed, and how deep are the cells.

After the first results of the NT analysis it was clear that the belt mount has exactly the same main structure as Alamannic examples analysed before: hollow, made out of 2 iron plates, the silver inlays about 1 mm deep. An interesting phenomenon is that the brass plates with copper cells and the silver plates all have more or less the same depth. The glass inlays are as deep as the thickness of the mount itself.

Ceremonial swords?

Two presumably ceremonial swords from the Netherlands were analysed. The first, the bronze sword or dirk from Jutphaas, dates from the Bronze Age, more precisely between the 15th to 14th century BC (figure 8). It is 42 centimetres long, rather thin and flat, the edges are not sharpened and it is perfectly symmetrical. It is believed to be a ceremonial sword of extra-ordinary high quality and without doubt a masterpiece of bronze working (Butler and Sarfatij 1970/1971, Fontijn 2001). The workshop where it was fabricated was presumably located in Brittany or South England, so it travelled over 800 km. The sword was found in 1947 during dredging activities for a harbour near Jutphaas. The finder, Giel Smidt, gave it to two boys who hung it on the wall of their bedroom. In 2004 the Dutch National Museum of Antiquities in Leiden acquired it.

In total there are five largely similar dirks known. Two originate from The Netherlands (Jutphaas and Ommerschans), two from France (Plougrescant and Beaune) and one from the UK (Oxborough). They are all very similar to each other and too large to handle and too thin to use. They are clearly ceremonial items (Butler and Fontijn 2007, 312). It is assumed that they all come from the same workshop. The Jutphaas dirk, however, is much smaller compared to the other four and because of that has been seen by some researchers as a falsification. Results from a NRCA analysis at the GELINA facility in Geel (Belgium) show that the composition of the Jutphaas dirk is very similar to two other dirks (Oxborough and Beaune see table 1). The upper part of the Beaune dirk differs in composition but this is a modern repair made from brass. The analysis demonstrates that, notwithstanding its size, the Jutphaas dirk belongs to the group of Ploughrescant-Ommerschans dirks and that the dirk is not a falsification. Moreover it demonstrates that all of the dirks were probably made in one workshop. This, in combination with their wide distribution and extraordinary shape and size demonstrates the special, symbolic character of these weapons.

Fig. 8 – A ceremonial sword from Jutphaas (The Netherlands) dating from the Bronze Age

	Ag	As	Co	Cu	Fe	Ni	Pb	Sb	Sn	Zn
*Oxborough**										
hilt	0.018	0.35	0.027	83.7	0.035	0.54	0.168	0.09	13.8	0.03
mid	0.017	0.36	0.028	85.8	0.040	0.54	0.148	0.10	13.9	<0.007
tip	0.017	0.34	0.029	84.4	0.036	0.53	0.170	0.10	13.5	<0.007
*Beaune**										
hilt **repair**	0.023	0.27	-	81.6	0.214	0.039	3.85	0.03	4.96	7.58
mid	0.019	0.23	0.026	84.6	0.023	0.544	0.138	0.10	13.6	0.012
tip	0.020	0.22	0.025	85.4	0.029	0.547	0.138	0.12	13.7	<0.007
Jutphaas										
top	0.0160	0.297	0.035	85.46	0.22	0.34	n.o.	0.106	13.50	0.106
tip	0.0124	0.226	0.033	86.07	--	--	n.o.	0.114	13.39	0.153

* from Needham (1990), n.o. = not observable

Tab. 1 – The compositions in weight% of three swords of the Plougrescant-Ommerschans type.

The bronze sword from Buggenum dates to the Middle Bronze Age between 1300-1100 BC (Butler and Fontijn 2007) (figure 9). It can be classified as a *Vollgriffschwert*, due to the solid grip, of the type *Vielwulstschwert* (Von Quillfeldt 1995), due to the multiple ribs. It is 68 cm long and its blade is 4 cm wide and was originally fabricated in Bavaria, Southern Germany or Austria. The sword does not appear to have been used; it has a fully decorated hilt (*cire perdue*-casting?) and a blade with a mid-rib and sharp edges. Based on these characteristics, it was labelled as ceremonial. The object is a very rare object within the Dutch Bronze Age assemblages. It was dredged from an old bedding of the river Meuse near Buggenum in the province of Limburg, the Netherlands in 1964. The finder, Mr. Peters, kept it in his possession until 1999 when it was purchased by the National Museum in Leiden.
The main questions for this object were if the hilt and the blade were produced separately or not and if the sword was a potentially functional weapon.

Three types of analysis were performed on the Buggenum sword, NRCA at the Geel Electron LINear Accelerator (GELINA) in Geel (Belgium) on 13 locations of the sword, neutron diffraction experiments on ENGIN-X of the ISIS facility of the Rutherford-Appleton Laboratory on comparable positions (Postma *et al.* 2010), and neutron tomography at FRM II in Munich. Figure 10 shows the sword at the GELINA facility in Geel in Belgium. Figure 11 shows some of the results from GELINA. It clearly demonstrates the differences in tin and copper weight ratios and the density between the blade (left part of the figure) and the hilt (right part). They are without doubt separately produced. A radiographic picture (figure 12) show the way blade and hilt are connected. This was confirmed by neutron tomography in Munich.

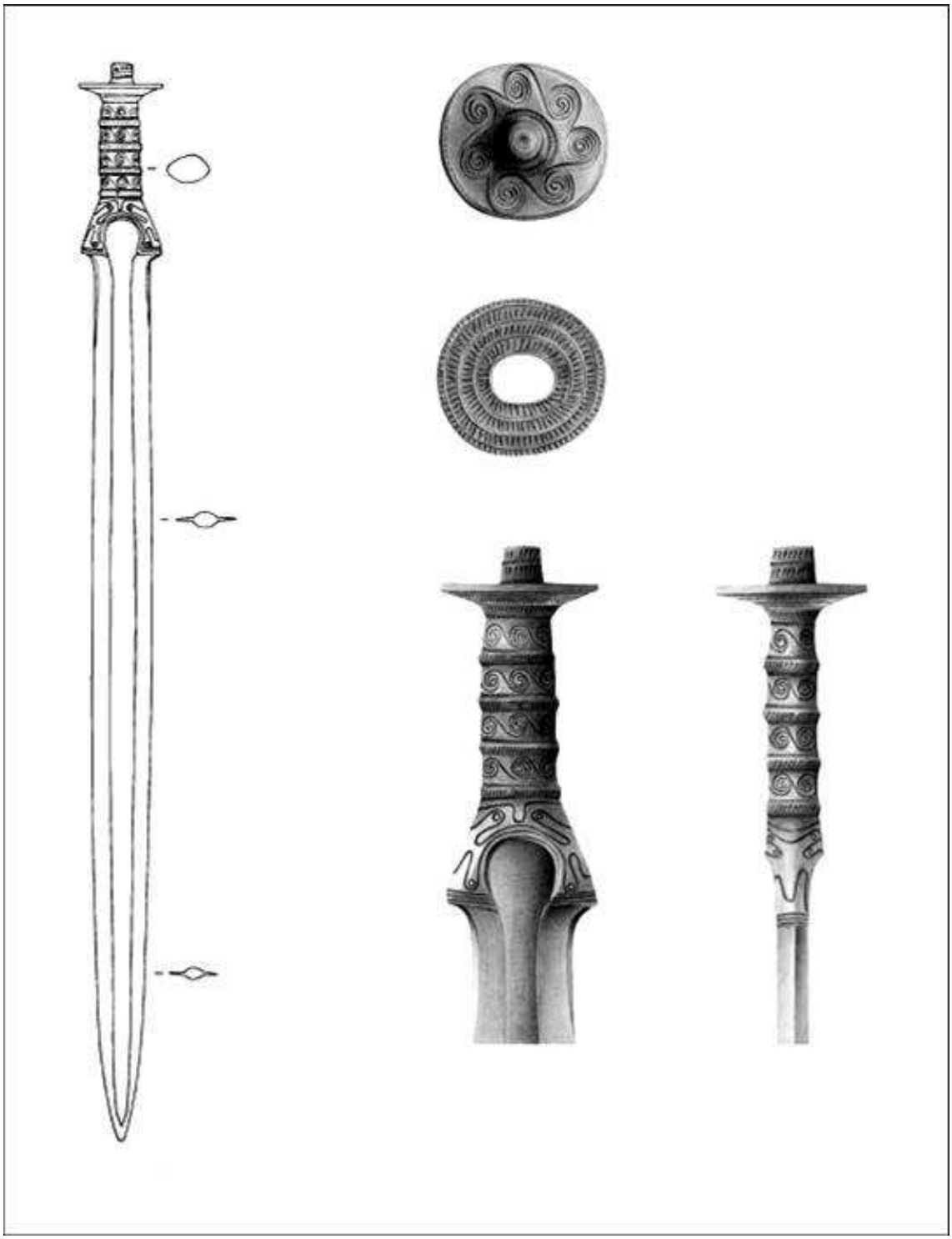

Fig. 9 – A bronze sword from Buggenum (The Netherlands) dating from the Middle Bronze Age.

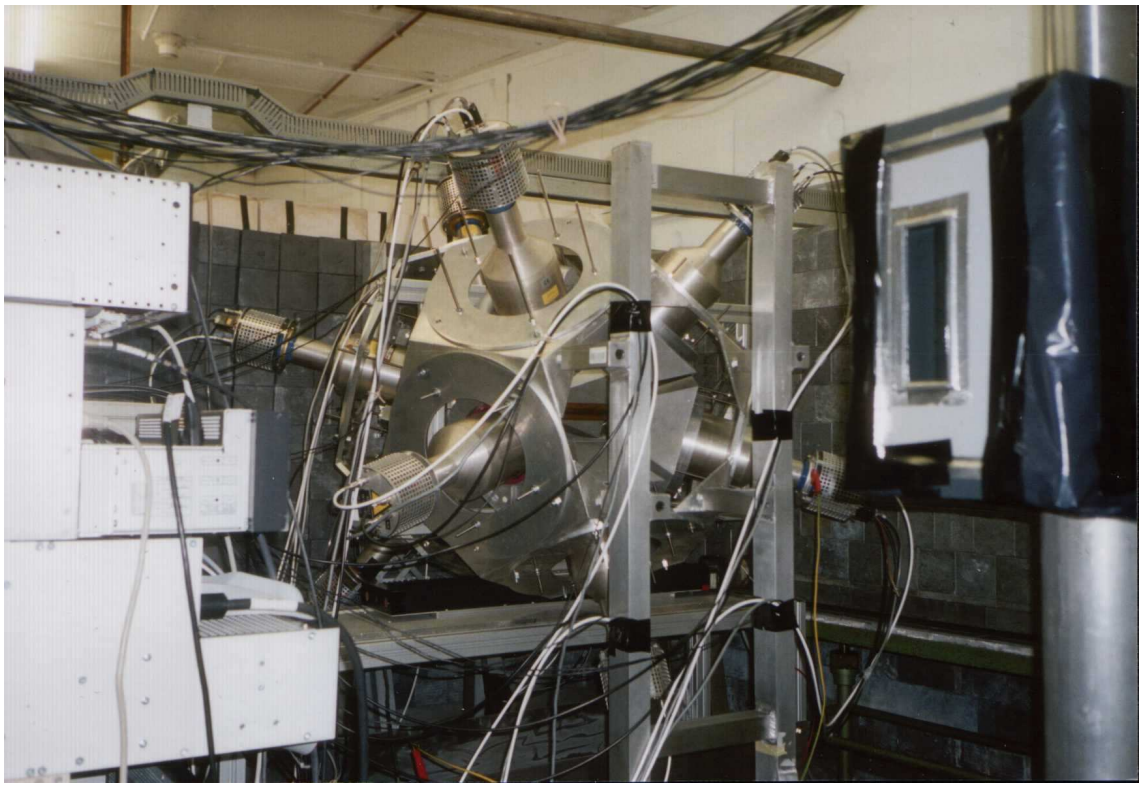

Fig. 10 – The Buggenum sword at the GELINA facility in Geel (Belgium).

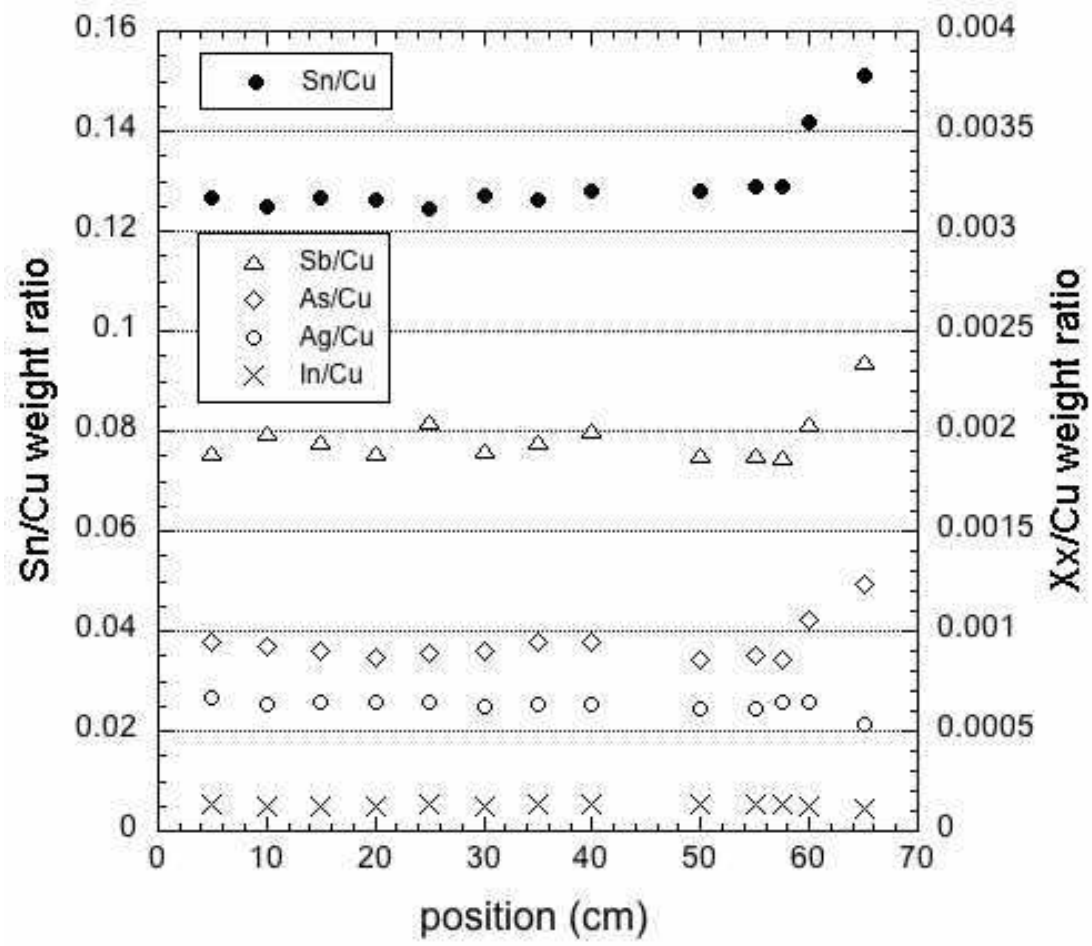

Fig. 11 – Results from the measurements in Geel (see text for details).

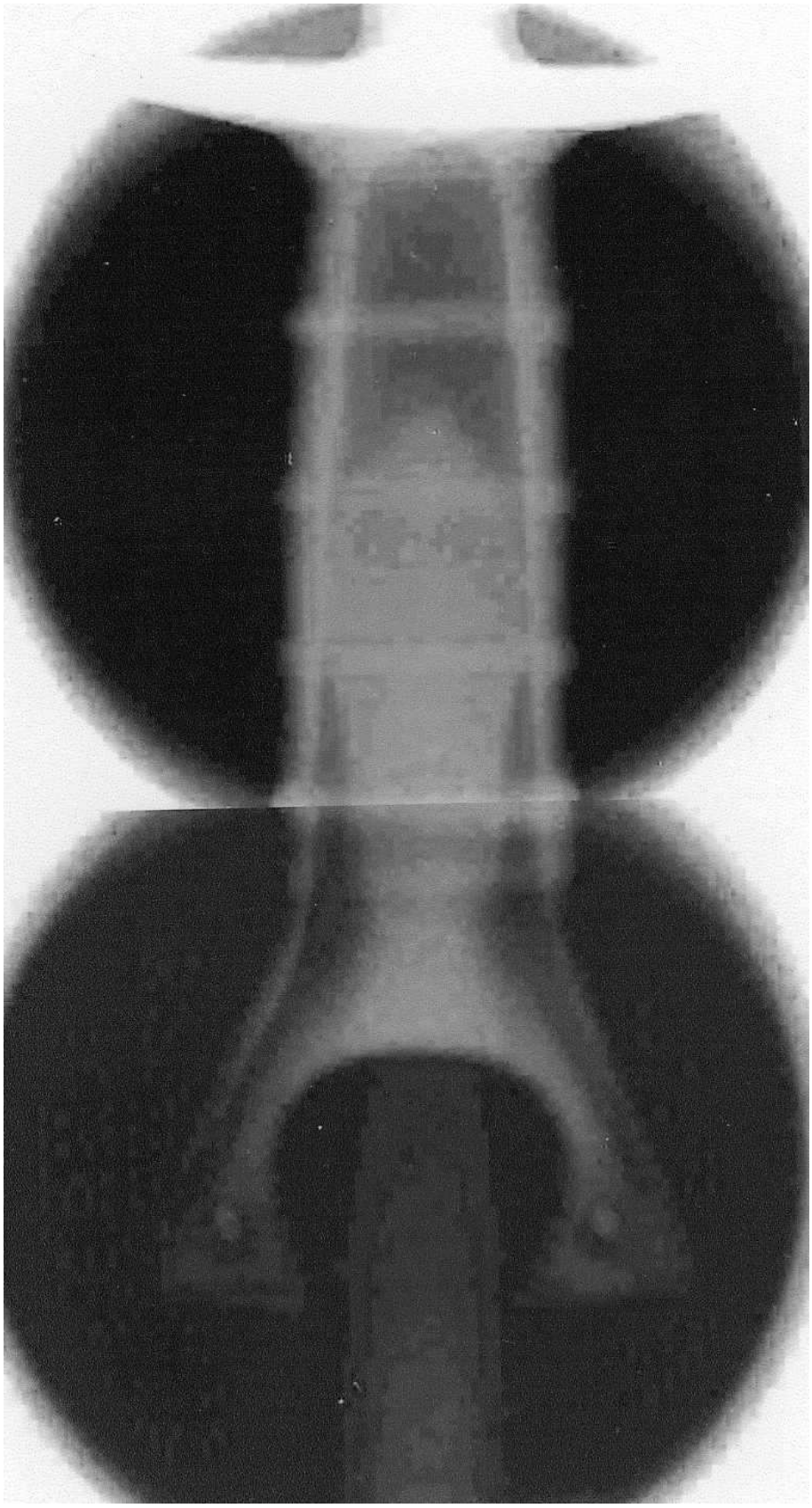

Fig. 12 – A radiographic picture from the Buggenum Sword.

Neutron diffractions and the Munich data furthermore indicate that the tin contents in the edges differ from the mid-rib of the blade. The edges are homogenized and hardened. The sword was thus manufactured as a potentially functional weapon. This casts some doubts on the label *ceremonial* with respect to the design of the weapon. In contrast to the Jutphaas dirk mentioned above, the Buggenum sword was not necessarily intended to be a ritual or symbolic object from the moment is was made.

Conclusions

The Ancient Charm project has been successful on all archaeological applications. It shows that the methods used can give answers for cultural heritage objects on topics like characterization (the Hungarian objects), provenance, technology, authenticity (Jutphaas) and functional or ceremonial use (Buggenum). It can also suggest methods for preservation or restoration (Ghiberti’s reliefs).

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