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NEUTRON RADIOGRAPHY AS A NDT METHOD IN ARCHAEOLOGY

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ABSTRACT

Applications of NR in archaeology are indicated when searching for the presence of materials containg hydrogen like organic materials wood, fabrics, leather, or corrosion products and by metals, in particular heavy metal lead. In the recent years the thermal neutron radiography (NR) has become a common non-destructive examination method at the National Museum of Slovenia, usefully complementing radiography by X- and gamma- rays and other NDT methods. The neutron irradiations are performed at the NR facility of the Ljubljana TRIGA Mark II research reactor at 250 kW power and using direct neutron imaging with thermoluminescent imaging plates (IP-ND's). Typical neutron fluences at the object plane are 10^6-10^7 n/cm^2, collected during rather short exposure times of few to 30 seconds and thus keeping the activation level of the object below of the level set by the safety standard for public release. In the paper several recent applications of NR in archaeology will be described. Objects examined by NR range from artefacts salvaged from river bottoms, small statues, bronze and iron age and medieval weaponry, bronze boat shaped fibulae, roman treasure found and even neolithic ceramic objects. Due to the enlargement of European Union and falling borders and availability of NR sources at several research reactors through the EU it is important to make aware the community of archaeologists, conservationist and restorers about the opportunities of NR in their field of research and applications.

Keywords: Archaeology, Conservation of cultural objects, Neutron radiography, Ljubljana TRIGA research reactor

1. Introduction

Non-Destructive Examination (NDE) techniques are of great importance for the examination of archaeological objects. Such objects studied by archaeologist are precious and often unique and destructive techniques should be applied exceptionally and only if non-destructive analyses do not give the results required and essential for the planned research.
Among the many NDE techniques available, capable of revealing the internal structure of objects, X-ray radiography (XR) became widely used and should nowadays be routine for the

study and/or the conservation of the majority of archaeological objects and other objects of art. The complementary thermal neutron radiography (NR) technique is far less used in this field. Until recently, the main reasons for this were the much smaller availability of suitable readily accessible neutron sources and/or the lack of efficient modern neutron imaging equipment. According to a recent monograph on the use of radiographic methods in the study of cultural objects, the drawback of NR could also be somewhat greater radiation hazards with neutrons, mainly associated with possibly induced, but short-lived, radio-activation of the object [1]. Nevertheless, the possibilities to use NR in the examination of cultural objects have greatly improved due to the recent development of small mobile or even portable neutron sources, and also the great accessibility of high performance stationary NR facilities (e.g. at research reactors), which can be attributed to greatly improved international scientific co-operation in Europe [2]. In addition, new efficient neutron imaging techniques enabling digital NR became commercially available [3, 4]. For this reason and to increase the awareness of archaeologists, historians and restoration specialists of the present possibilities of NR in their fields of interest it seems worthwhile to review some applications of digital NR to archaeological objects carried out at the Ljubljana TRIGA Mark II research reactor of the J. Stefan Institute (JSI). In this paper the radiographic characteristics of NR relevant to archaeological applications are summarized. The archaeological objects, studied by the staff of the National Museum of Slovenia, were excavated in the ground or found in a river. The non-destructive examinations were part of conservation procedure, or needed to draw conclusions on the manufacturing technology, or to give a clue to their function.

2. NR in examination of archaeological objects and other cultural items

The use of NDE techniques in revealing the internal structure and composition of an object is already important in the planning stage of its conservation. During the process of conservation and afterwards, NDE of the internal structure of objects is very useful for evaluation and verification of the conservation procedures applied. Further, NDE of the object's interior is extremely important in the study of ancient manufacturing technologies and in providing a clue to their function. The examination and characterization of internal structure and composition can be difficult task, in particular for excavated archaeological objects, as these can be covered by soil, sand or mud, often in a mixture and, when metals are present, also mixed together with various corrosion products. NDE can be a complex task since the object, its parts or contents can consist of various materials: metals, inorganic components such as stone or ceramics and organic materials, such as wood, fabrics, leather, food remnants, fragrances etc.

Several NDE methods can be used for the examination of the object's interior, but only the application of various complementary NDE techniques can provide the maximum information. X-ray or even Gamma-ray Radiography (XR) and Neutron Radiography are non-destructive techniques which usefully complement each other. Principles of NR, equipment and typical applications are described elsewhere [5]. XR shows only density variations and has some other drawback in archaeological applications, particularly as the more durable metals such as gold, silver and lead are nearly opaque to X-rays. On the other hand, NR is quite sensitive in detection of hydrogenous materials such as water, water-logged ceramics, organic materials such as wood or water-logged wood, plants, seeds, food remnants, leather, textiles, paper, fragrances, tar, epoxy resins etc. Technical metals such as copper, tin, iron, bronze, as well as lead, are fairly transparent to neutrons. This enables to uncover fine structures inside thick metal layers which are not accessible for x-ray radiography. Non-metals (for example rock materials, minerals, glass, ceramics) are also more or less transparent to neutrons. With NR, for example, it is possible to visualize hydrogen-containing materials inside metal artefacts much better than with X-rays, whereas the XR technique is more suitable for visualizing the integrity of metal objects or metal parts of objects made of organic materials.

A variety of NR examinations of archaeological and other cultural objects have been performed at JSI in the past:

1. Cast metal (iron, bronze) objects such as jars, statues and brooches were inspected for remnants of clay moulds to elucidate the production technology or for corrosion damage [6];
2. Metal objects such as closed vessel, an iron reliquary cross, handle of a medieval judicial sword and gun ammunition were inspected for their contents, including remains of organic materials such as wood ,bones, strings or gun powder respectively [6,7];
3. Unearthed metal objects, for example ancient hand weapons, buckle belts and tools were examined for the presence of wood, leather or other fabrics in their internal structure and to study their production technology [8,9];
4. Restored metal, stone or ceramic objects were inspected for the presence of adhesives, glued parts, epoxy filler material or epoxy protective coatings [6, 9].
5. Pottery objects are not suitable for NR since the constituent elements give no great contrast, unless at least traces of boron are present in the glaze coating. However, recently a Roman treasure find, a large ceramic pot, filled with soil and sand, was successfully examined in order to characterize the internal artifacts and to localize their position. This work is described in more detail in a companion paper at this conference [10].

3. Experimental requirements

Potential users should be aware that the equipment needed for the NR examinations is basically different, usually more expensive and more demanding for operation than equipment for common XR, and consists of:

1. A neutron source together with a neutron collimator and neutron/γ-ray filters, neutron beam shutter, neutron shielding and radiological protection equipment, and possibly with some remote object handling equipment;
2. Equipment for neutron image detection and processing;
3. Equipment for verification of the radioactivation of the object after neutron exposure, possibly also enabling identification of radionuclides in the sample by means of high resolution gamma spectrometry (INAA).

Typical neutron radiography facilities with neutron sources and neutron imaging equipment are well described elsewhere, e.g. [5]. Currently there are about 15 NR facilities based on research reactor neutron sources and several neutron generator based NR facilities operating in Europe alone [2]. Therefore the opportunities to perform NR in Europe are not insignificant, in particular in view of the presently increased scientific co-operation between European countries in general. Several NR facilities exist at various beam ports of the world-wide very popular Gulf General Atomic TRIGA research reactors, e.g. in Ljubljana (Slovenia), Pavia (Italy), Vienna (Austria). Most of these NR facilities are equipped with modern neutron imaging systems of high performance and enabling digital NR. At a few EU research institutes Computed Neutron Tomography (CNT) is available [EC COST 524 project, 2002].
However, the operator of the NR facility should consider some important wishes of the potential customer, i.e. archaeologists, conservators, historians:

- The neutron exposure as well as the total exposure of the artifact should be as low as possible to prevent or minimize radiation damage and induced short-living radio-activation of the object. In most cases the object should be returned to the customer immediately after examination and only exceptionally will the customer allow the object to stay in a safe storage room for some time to allow the radioactivity to decay to an acceptable level. The level of specific induced radio-activity should be below the standard clearance level of ~75Bq/g, required for the release of a "non-radioactive" sample. The use of neutron beams of moderate intensity and/or short enough neutron exposure times are thus indicated.

Neutron beams of lower intensity should be used in combination with high performance neutron image detectors, e.g. image detection systems based on intensified CCD cameras, photo luminescent imaging plates or amorphous silicon detectors.

- Neutron beams of large useful area at the detector plane are preferred to avoid multiple neutron exposures.
- The image quality (image sharpness, detail discernment) should be comparable to that of XR. Typical performance and image quality characteristics of some advanced image detectors used in digital XR and NR are described elsewhere [4,5].

4. NR at Ljubljana TRIGA Mark II research reactor

The NR facility in the thermal column of the Ljubljana TRIGA Mark II research reactor has already been described previously [6]. It has been used successfully for the NR examination of archaeological objects in the past [7-9]. The intensity of the collimated neutron beam is rather weak ($3.5 \; 10^5$ n/cm^{2}s) and its characteristics match to a significant extent the characteristics of the mobile neutron generator based NR neutron sources [2]. The rather slow film/Gd screen based direct neutron high imaging technique was efficiently substituted by fast direct neutron imaging using Gd doped imaging plate neutron detectors (IP-NDs) produced by FUJI Photo Film Co.. The IP-NDs are read out by a FUJI BAS 1500 reader with 0.1mm pixel size and 10 bit digitalization. The use of FUJI IP-NDs reduced the exposure time from 1.5 h with the Gd screen/radiographic film method to only 2-100 seconds (factor 100), greatly increasing the speed of the NR non-destructive examinations and reducing the activation of the object. The pixel size of other modern, currently commercially available IP readers is of the order 0.025 mm which is very near to the inherent resolution limit of the very slow direct neutron imaging with thin Gd metal screens and fine grained single coated radiographic films.

5. Examples of recent applications to archaeological objects at the National Museum of Slovenia

Recent NR examinations of some objects belonging to the cultural heritage, performed at the relatively weak neutron beam of the Ljubljana TRIGA Research Reactor NR facility of J. Stefan Institute are reviewed below. They illustrate new possibilities of digital NR in the examination of cultural objects, brought about with the introduction of new neutron image detection systems.

5.1 Sword of the municipal judge of the city of Ptuj, dated to the 16th century A.D.

The sword, photography presented in Fig. 1a, is kept in the Ptuj Regional Museum and was studied by the National Museum of Slovenia where conservation was also carried out. The objective of NR was to examine the construction of the hilt with a iron pommel, in particular how the hilt covered by a gilded silver sheet, decorated by engravings, was fixed to the tang of the sword made of steel. NR, presented in Fig.1b, showed that two wooden linings enclosed the iron tang. They were bound together with cord - a detail which could not be observed by XR. The NR examination also revealed that an organic material - perhaps a resin used as glue - probably filled the hollow space around the tang in the iron pommel.

5.2 Iron wheel lock pistol, 16th century A.D.

This iron pistol was found almost intact in a river in Slovenia. Its metal parts were very well preserved but the wooden parts were absent. NR examination (Fig. 2a) of the pistol showed the very well preserved wheel lock and a quartzite stone. NR examination of the barrel (Fig. 2b) indirectly revealed the lead bullet and that the barrel was loaded with gunpowder. The position of the lead bullet in the barrel is indicated indirectly by the round shape of the mixture of mud and corrosion products or gunpowder deposited around the bullet, since lead is almost transparent to neutrons. The lead bullet alone was clearly detected by XR, but there was no hint of remnants of gunpowder (Fig. 2c). The lead bullet was slightly flattened, apparently when it was pushed inside and against the gunpowder during loading.

(a) (b)

Fig. 1: A medieval sword of municipal judge, photography (a) and NR (b). NR reveals that the pommel of the sword is made of two wooden parts and an iron tang, fixed together by a string (marked in the figure) which was not possible to observe by XR.

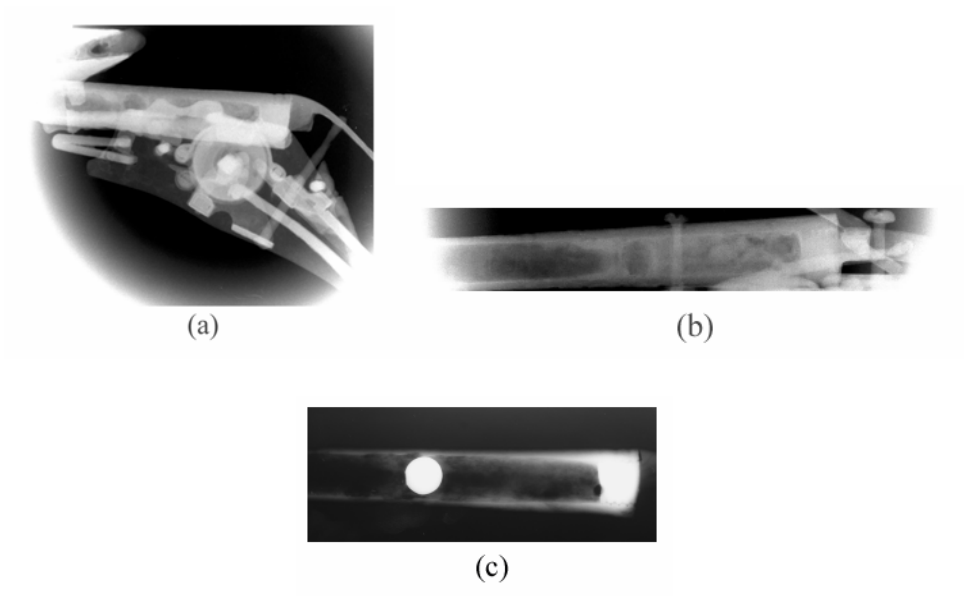
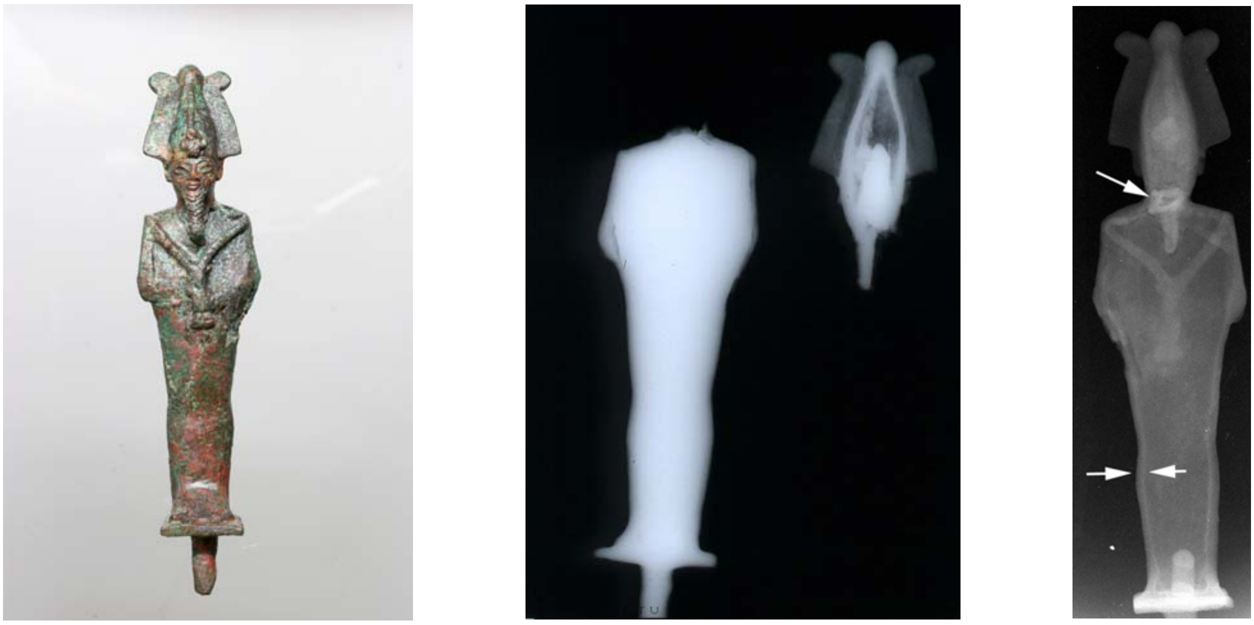


Fig. 2: NR and XR of a wheel-lock pistol: Wheel-lock mechanism, quartzite stone and indications of the presence of gunpowder can be observed in the NR photographs of the pistol (a) and the barrel (b). The lead bullet can not be directly observed by NR. Its presence in the barrel is revealed by the detection of the remnants of mud, corrosion products and gunpowder clogged around the bullet (b). The position of the lead bullet is clearly revealed by the XR (c).

5.3 Bronze statuette of Osiris, dated 8. century B.C.

The bronze statuette of Osiris from Thebe was found with the head broken and separated from the body. The photo after the restoration is presented in Fig. 3a. The head was attached to the body by an organic glue during the restoration and which is seen on NR (Fig.3a). The statuette was made by bronze casting in a ceramic mould, which was later removed. Some ceramic sand still remained in the upper part of the head and which can be observed as a cavity in XR before the restoration. (Fig. 3b). In XR the internals of the statuette are obscured by lead, with which the statuette was filled after the casting. The XR shows the blurring outline of the lead and the bronze mantle can not be seen. On the contrary the thickness of the bronze mantle and other bronze parts (hands, royal insignia – cobra, whip, sceptre) can be easily observed by NR (Fig. 3c). The lead is almost transparent for neutrons and opaque fore x-rays.



(a) (b) (c)

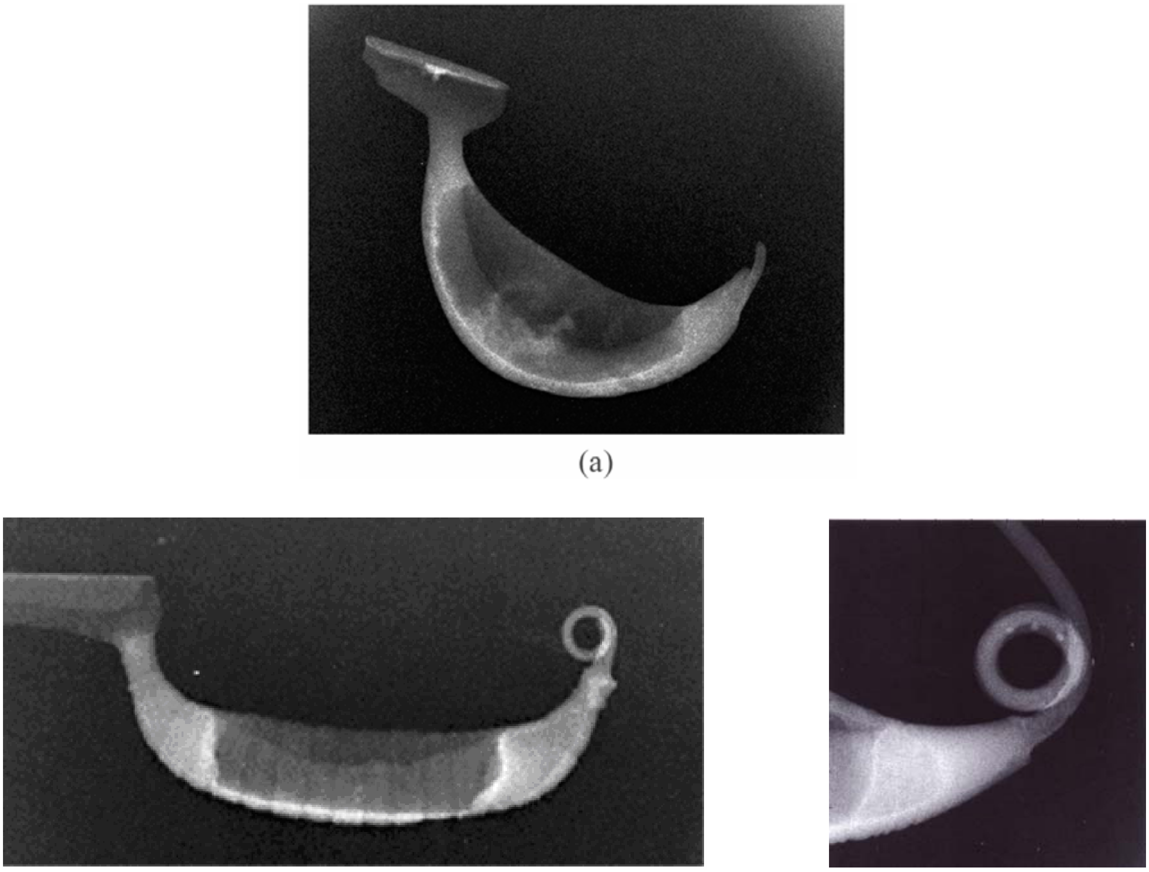
Fig. 3: Photography (after the restoration) on (a), XR (b) and NR (c) of a bronze statue of Osiris. The mantle of the statue is made of bronze metal and the internals are almost completely filled with lead. The NR reveals the thickness of the bronze mantle, some cavities without lead and also a place, where the head was fixed to the body by organic glue for restoration. Cavity in the head without lead and filled with ceramic mould sand is clearly observed by XR.

5.4 Bronze boat-shaped brooches (fibulas), 7th century B.C

The brooches (fibulas) are kept in the National Museum of Slovenia where they were studied and conservation was carried out. Several bronze boat-shaped brooches (fibulas) from different archaeological sites in Slovenia were inspected both by XR and NR. Interesting features detected by NR are the remnants of clay moulds, which were not completely removed after the casting (Fig. 4a, b). In a few cases when the brooches (fibulas) were drilled to collect material for chemical (ICP) analysis, the holes filled by epoxy resin were clearly seen by NR as well.

5.5 A neolithic ceramic rattle box, dated 2200-3000 B.C.

The neolithic rattle box was excavated already in 1875-1877 on the location of Dežman's crannog on the Ljubljana bogland near village Ig and was never opened. Its internals are therefore orginal. The details of the internals- rattle stones, made either of ceramic or stones, are observed. The rattle box is made of two parts and its closure can also be seen. The constituing elements of ceramics (Si, Ca, Mg, Na) are transparent for neutrons and the attenuation of neutrons and the contrast are due to the hydrogen (moisture, crystal water). More data will be provided by XR.



(a) (b) (c)

Fig. 4: NR of a boat-shaped brooches (fibulas) (7th century BC): The shape of the remnants of the ceramic mould can be seen inside the brooches (fibulas) (4a and 4b). Light areas on the image correspond to smaller neutron exposure and hence to more neutron opaque parts containing ceramic material. Three holes drilled to collect material for chemical analysis and filled by epoxy resin can be also seen in upper part of Fig. 4c.

Fig. 5: NR of a neolithic ceramic rattle box (2200-3000 B.C.) with rattle balls made either of stones or ceramics.

6. Conclusions

The introduction of modern neutron image detectors of high performance and the improved accessibility to stationary NR facilities around Europe has made NR attractive for applications in archaeology, for conservation of archaeological objects and other objects belonging to the cultural heritage, as well as for the study of ancient manufacturing techniques. NR as a complementary method to the more common XR found its place as a regular and suitable NDE method in the National Museum of Slovenia. NR is unique in examination of organic or metal parts and artefacts through thick layers either of a heavy metal lead or non-metals as soil, sand or ceramics.

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