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NEUTRON IMAGING STUDIES WITHIN THE »neu_ART« CULTURAL HERITAGE PROJECT

The importance of neutron radiography in archaeology and in cultural heritage is assessed by now considering that the first radiographies with neutrons were already performed in the seventies (Hilling 1975). However, the recent development of new methods, detectors and instruments and the availability of neutron radiography at smaller reactors has allowed researchers to obtain more and more interesting results. The aim of the this work, car ied on within the neu_ART project, has been to determine the imaging potentials of neutron radiography and tomography of artistic objects, in particular those containing metallic artefacts performing several measurements at two laboratories, the NECTAR facility at the FRM II reactor of the Technische Universität München (Germany) and the INES facility at the ISIS spallation source of the Rutherford Appleton Laboratories in the UK.

The goals of the measurements were:

– the evaluation of the performance of the imaging systems in terms of linearity, spatial resolution and dynamic range, together with the dependence of these quantities on the detection system;
– the study of the attenuation of the beam intensity as a function of the target thickness for different materials and the comparison of the neutron penetration capabilities with thermal and fast neutrons;
– the analysis and the interpretation of radiographs and tomographs of some few artistic objects in order to assess the obtainable information with these techniques.

In the following, after the description of both apparatus, the main results will be presented.

Materials and methods

The INES and NECTAR facilities

INES, the Italian Neutron Experimental Station (Celli et al. 2006; Grazzi et al. 2007), is an apparatus designed for material investigations through diffraction measurements with thermal and epithermal neutrons at the ISIS neutron source of the Rutherford Appleton Laboratories (UK). Neutrons are produced by spallation through the collision of a pulsed proton beam on a tungsten target and slowed down to the energy range 7.8 meV to 5 eV. The cross section of the INES beam, at the sample position, is a square of about $4 \times 4\,cm^2$ size with a uniform intensity of 1.1×10^7 neutrons/$cm^2 \times s$ and an L/D ratio of about 90. The INES facility features a mechanical instrumentation, used to translate and rotate the test object, and a detector, positioned at a distance between 1 and 10 cm from the object, composed by a scintillator described in the following section and a CCD camera. The latter (The Imaging Source DMK21BF04) is a low price commercial product because the radiation damage imposes frequent replacements. It is segmented in 640×480 pixels, each corresponding to a region of about $100 \times 100\,mm^2$ of the image field, and works at room temperature. The CCD is read out through an 8 bit ADC, leading to a nominal dynamic range of 256 grey levels; the maximum integration time is 30 s. To evaluate possible limits of this camera, a second CCD camera with a larger nominal dynamic range (12 bit) and with the possibility to set wider time intervals for the signal acquisition (Allied Vision Technology Manta G-032B) has been used.

NECTAR, the Neutron Computerized Tomography And Radiography facility (Bücherl/Lierse von Gostomski 2006; Bücherl/Guo/Lierse von Gostomski 2009), has been designed specifically for radiography and tomography with fast neutrons at the FRM II research reactor of the Technische Universität München (Germany). Fast neutrons are produced by fission in two uranium enriched converter plates positioned at about 1 m of distance from the reactor core inside the moderator tank and can be collimated to reach an L/D ratio of about 230. Moreover two filters made of cadmium and lead are added to absorb respectively thermal neutrons coming from the reactor and high energy photons produced by the fission process. The area of the beam transverse section at the sample position is $37 \times 31\ \mathrm{cm}^2$ in maximum with an average intensity of 5.4×10^5 neutrons/$\mathrm{cm}^2 \times \mathrm{s}$ and about 1.7 MeV mean energy. A sample manipulator allows the translational, rotational and vertical positioning of the test object. The detection system consists out of a scintillator screen in combination with a CCD camera. The latter is a low noise ANDOR DV434-BV camera, originally developed for astronomy applications, and cooled by Peltier cells. It is segmented in 1024×1024 pixels, each of them corresponding to $13 \times 13\ \mathrm{mm}^2$ of the image field. The nominal dynamic range is 16 bit and it is possible to integrate the signal up to a time of 2000 s corresponding to the saturation level.

Properties of the analyzed scintillators

One of the main studies performed at the two neutron facilities has been the comparison of different scintillators. They have been properly selected based on the properties described in the following, with the aim of highlighting their detection potential for this kind of application.
In the following the main properties of the tested scintillators are reported.

INES

Thermal neutron screen NDG (Applied Scintillation Technology). It is composed by ZnS(Ag) mixed with lithium fluoride ^{6}LiF with a relative weight of 2:1. Two versions of this scintillator have been tested. The scintillation material in the first type is inserted in a plastic matrix of 450 µm thickness while in the second one is deposited on a thin aluminum layer of 1 mm with a total thickness of 250 µm.

NECTAR

ZnS(Ag) in polypropylene matrix, thickness 2 mm. This is the neutron converter actually used at the NECTAR facility. It has been characterized in order to have a reference for the other scintillators. The scintillating material is ZnS(Ag) and the luminescence is due to charged particles generated by the interactions between neutrons and nucleus of the polypropylene matrix elements.
Thermal neutron screen NDG (Applied Scintillation Technology). It is the scintillator ZnS(Ag): ^{6}LiF embedded in plastic matrix, 450 µm thick, used at INES. Since it is compos ed by the same scintillation material as that of the main NECTAR converter screen but being thinner, it allows to check the dependence between resolution and scintillator thickness with fast neutrons.
Plastic scintillators (EJ200 EJ212 Eljen Technology). Two converter screens have been tested, both with a thickness of 20 mm. The first one, EJ200, is solicited as the best general purpose organic scintillator because it detects almost all types of radiation with a high converter efficiency. The second, EJ212, is the plastic scintillator mostly used for fast neutron detection in nuclear physics. Unlike the EJ200 screen, it is not sensitive to high-energy gamma rays. This makes it more suitable for applications where the neutrons to detect are in a mixed neutron-gamma field, as at the NECTAR facility.
Agfa Curix Ortho Regular X-ray converter screen. It is composed by gadolinium oxifluoride activated with terbium (Gd_2O_2S:Tb), an inorganic scintillator, and commonly used as support screens in the traditional X-ray radiography. It is available at a moderate price, also in big sizes. This fact, together with the small thickness of 1 mm, makes it particularly indicated for an in-depth characterization at the NECTAR facility.

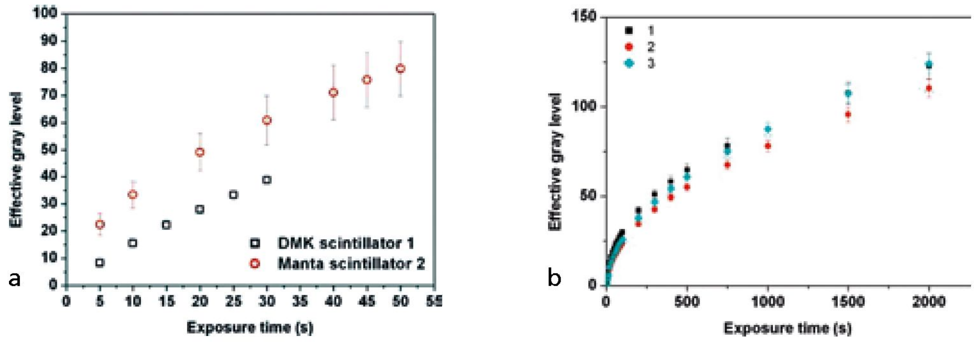


Fig. 1 Number of effective grey levels as a function of the signal integration time for different combinations CCD-scintillator. – **a** results of INES (1 = Thermal neutron screen NDG 250 mm; 2 = Thermal neutron screen NDG 450 mm). – **b** results obtained at NECTAR (1 = ZnS[Ag] pp matrix thickness 2 mm; 2 = plastic scintillator EJ200; 3 = plastic scintillator EJ212). – (Graphic by the authors).

Results

Line arity

Open beam images, with increasing exposure times, have been acquired at INES and NECTAR. They have been used to study the linearity of all the available imaging systems composed by different combinations of scintillator and camera. The mean of grey levels (signal) and the corresponding standard deviation (noise) have been computed inside a uniform area of 10 × 10 pixels, using the assumption that the signal fluctuation among 100 consecutive pixels of an image is equal to the fluctuation of grey levels of a single pixel for a sequence of 100 images.

The results indicate that at INES, by using the DMK21BF04, the 450 mm thickness scintillator and the maximum integration time of 30 s, it is possible to exploit only half of the camera nominal dynamic range. On the contrary the camera MANTA G-0328, combined with the same scintillator, allows to reach the saturation when the signal is acquired at the maximum exposure time of 60 s. For all the scintillator-camera pairs the linearity between the signal and the exposure time is verified up to the maximum grey level. The highest slope has been achieved with the thicker scintillator (DMK: 3.77 ± 0.03 grey levels/s; MANTA 72.4 ± 0.2 grey levels/s) which features the best neutron conversion efficiency.

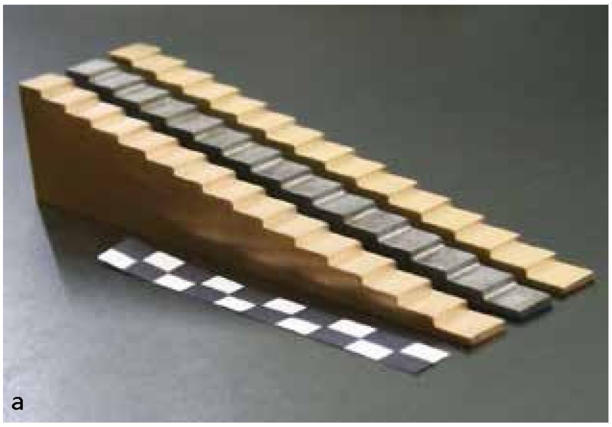
At NECTAR open beam images have been acquired at different exposure times (from 0.1 to 2,000 s) by combining the ANDOR camera with all the scintillators described in the previous section. For all the combinations, the signal showed to be linearly dependent on the exposure time and the maximum angular coefficient (8.82 ± 0.04 grey level/s) has been achieved with the 2 mm thickness scintillator of ZnS(Ag) in polypropylene matrix.

Dynamic range

The signal-to-noise ratio as a function of signal, in all the imaging systems, resulted to be described by a power function with a fractional exponent, indicating that the main contribution to noise originates from statistical fluctuations on the number of photons hitting the camera. Indeed, if the assumption of signal fluctuations following a Poissonian statistics is made, it should follow the functional form $y = A \cdot x^{0.5}$. Moreover the effective dynamic range (L), i. e. the number of grey levels which can b e effectively distinguished taking into account the noise contribution, has been computed for all the camera-scintillator combinations according to the method described in (Betuzzi et al. 2007). L is related to signal (S) and noise (N) by the equation:

$$L = \int_a^b \frac{1}{N(S)} dS$$

where a and b are respectively the highest and the lowest limit of the available signal range. Compari-



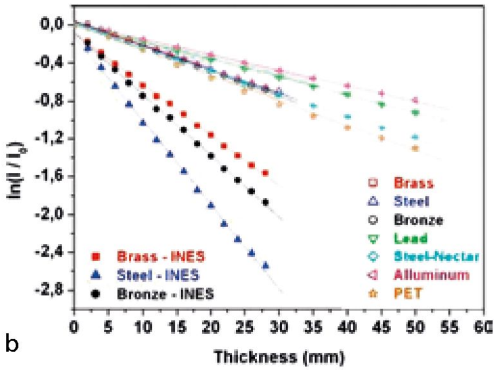


Fig. 2 **a** bronze, steel and brass step wedges. – **b** relative beam intensity as a function of the step thickness obtained at INES with the imaging system composed by the MANTAG-032B camera and thermal neutron screen NDG 450 mm thick scintillator and obtained at NECTAR with the ZnS(Ag) pp matrix scintillator 2 mm thick. – (Graphic/photo by the authors).

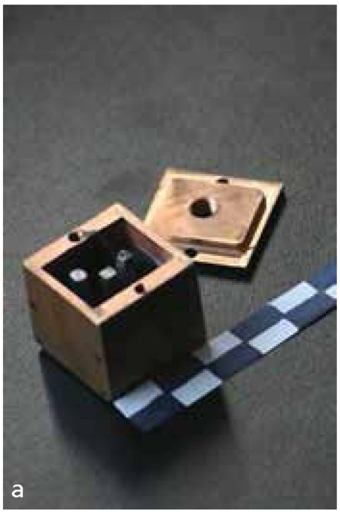
sons between the different imaging systems have been performed with the same exposure time. At INES the highest dynamic range (80 effective grey levels) corresponds to the combination MANTA camera – 450 µm thickness scintillator while the lowest value corresponds to the combination DMK camera – 250 µm thickness scintillator (**fig. 1a**). At NECTAR the ZnS(Ag) scintillator in polypropylene matrix with a thickness of 2 mm and the two plastic scintillators EJ200 and EJ212 have a similar behavior achieving respectively 123 ± 7, 110 ± 5 and 124 ± 6 effective grey levels for exposure times of 2,000 s each (**fig. 1b**).

Spatial resolution

The spatial resolution, expressed in terms of Edge Spread Function (ESF) and Modular Transfer Function (MTF) (Hall 1979), has been computed for all the imaging systems by using the image of a sharp edge. At INES, the best spatial resolution (182 ± 11 µm, 4.0 ± 0.2 lp/mm) has been obtained with the DMK camera combined with the thinner scintillator of 250 µm. At NECTAR the best spatial resolution (659 ± 7 µm, 1.11 ± 0.01 lp/mm) has been achieved with the thermal neutron screen NDG of 450 µm thickness; this value is larger than the resolution obtained at INES with the same scintillator. In fact, although the spatial resolution is related to the L/D ratio (the higher the L/D, the lower the spatial resolution) and this ratio is higher at NECTAR, each pixel at INES corresponds to 100 × 100 mm^2 of the image field compared to 300 × 300 mm^2 at NECTAR. By using the 2 mm thickness ZnS(Ag) scintillator in polypropylene matrix, it is possible to resolve at NECTAR details of 900 µm; however, this spatial resolution worsens up to 2 mm with the two plastic scintillators because of their large thickness.

Elemental separation

Finally, the characterization has been completed with neutron radiographs of step wedges made out of bronze, brass, steel, lead, aluminum and PET manufactured on purpose for this analysis. For all the combinations camera-scintillator the beam intensity attenuation has been studied as a function of the thickness of material. **Figure 2** shows the results obtained at INES and NECTAR. In both cases the experimental data follow the exponential attenuation described by the Lambert-Beer law, the main difference being the maximum beam penetration depth which is higher at NECTAR (50 mm), and the different elemental separation potential. This can be



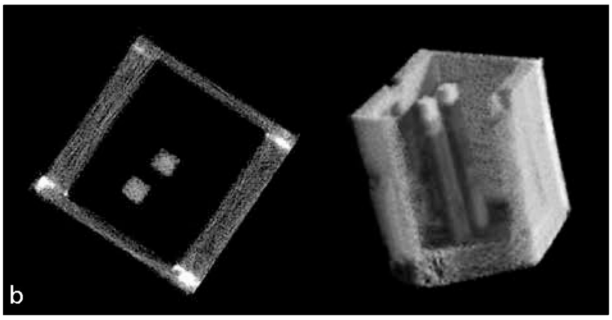


Fig. 3 **a** photo of the cube. – **b** reconstruction of a horizontal section (right) and 3D rendering (left). – (Photo F. Grazzi).

clearly observed at INES where the different beam attenuation of bronze, brass and steel is clearly evidenced by the angular coefficients of the straight lines which represent, in logarithmic scale, the exponential behavior of the beam attenuation as a function of the material depth. These straight lines are hardly distinguishable for some of these materials at NECTAR.

Tomographies

Given the increasing interest for neutron tomographic applications, a bronze cube of 25 mm side length (**fig. 3a**) has been realized with faces of different thickness, ranging from 4 mm to 2 mm which correspond to the typical thickness of bronze statues. Moreover two bronze rods and one of aluminum with square section of $3 \times 3\,mm^2$ and 15 mm height, have been inserted inside the cube together with two circular drills of 1 mm diameter in its thinner face to simulate respectively the presence of internal structures and metal casting air bubbles. The tomography of the cube has been performed at INES with the DMK camera and the thermal neutron screen NDG of 250 mm thickness. The cube has been positioned on a rotary stage and the angular step width has been set to 0.7°. Images have been acquired with an exposure time of 16 s. The tomographic reconstruction has been performed using the Filtered Back-Projection (FBP) method, implemented into the Parrec software developed by the University of Bologna (Brancaccio et al. 2011). In **figure 3b** horizontal section of the cube and the 3D rendering are shown. It is clear from the reconstructed section that it is possible to resolve details of a millimetric scale and to locate all the metallic structures inside the cube. Only the aluminum rod is not easily visible because of the aluminum cross section which is about one order of magnitude lower than the steel one.

Finally, a bronze hand (**fig. 4**) has been prepared at the Centro di Conservazione e Restauro »La Venaria Reale«/Italy, using ancient techniques of metal artifact casting, to evaluate the neutron tomography potential on works of art. The tomography of this object has been realized at NECTAR by acquiring 180 projections with an angular step width of 1°. Each projection has been obtained by the sum of 4 images with 60 s exposure time each, for a total acquisition time of 13 h. Results are reported in **figure 4** where the 3D rendering and three different sections (axial, transverse and horizontal) are shown. Neutron tomography allowed to reconstruct the width of the external shell and some internal structures as the presence of metallic filaments of millimetric size and low density materials which compose the casting sand.

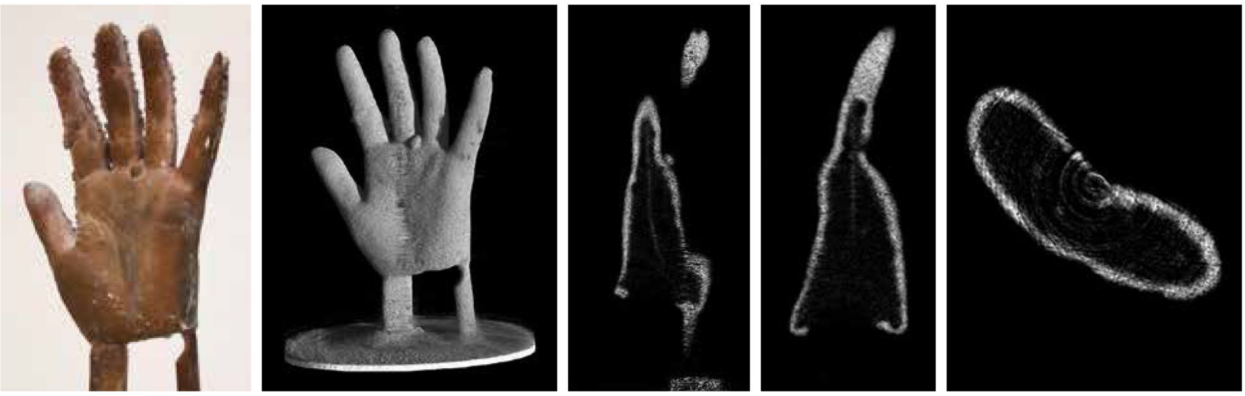


Fig. 4 From left to right: picture of the hand, 3D rendering, axial, transversal and horizontal sections. – (Photos by the authors).

Conclusions

The analysis of the imaging capabilities of the NECTAR beamline at FRM-2 reactor and INES beamline at ISIS spallation source were performed to evaluate linearity, dynamic range, spatial resolution, elemental sensitivity and tomographic capability with interesting results in terms of reliability of the methods and quality of the results. The large difference between the flux of these beamlines compared to the one of a compact source makes the latter unadapted to be used to provide neutrons to a neutron imaging compact beamline.

Acknowledgements

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Summary / Zusammenfassung

Neutron Imaging Studies within the »neu_ART« Cultural Heritage Project

The potential of imaging by thermal and fast neutrons has been assessed in the Cultural Heritage field, given the fact that this technique is non-invasive and neutrons can penetrate many layers of thick objects. This method provides valuable information to assist preservation and restoration activities and to help dating or attributing through the understanding of the manufacturing techniques. An investigation of the imaging capabilities of the Italian Neutron Experimental Station INES (at the ISIS pulsed neutron source of RAL laboratories, UK) and the Neutron Computerized Tomography and Radiography laboratory (NECTAR at the FRM II reactor in Munich) applied to artwork objects made of metal alloys was carried out. For this purpose custom samples have been prepared, following size and composition of typical ancient works of art. Through neutron tomography the internal structure of the testing metal objects was reconstructed and millimetric scale details inside them were resolved. Moreover, several measurements have been performed to fully characterize the performance of both apparatus. Their imaging response has been analyzed in terms of linearity, signal to noise ratio, dynamic range and resolution by the usage of scintillator screens of different chemical composition and thickness. A detailed description of these studies is presented.

Neutronenbildgebung-Untersuchungen innerhalb des »neu_ART« Kulturerbe-Projekts

Untersucht wurde das Potenzial von Bildgebung mit thermischen und schnellen Neutronen für das Gebiet des Kulturerbes, da diese Technik nicht-invasiv ist und Neutronen viele Schichten dicker Objekte durchdringen können. Diese Methode liefert wertvolle Informationen, die bei der Konservierung und Restaurierung hilfreich sind und durch das Verständnis der Herstellungstechniken eine Datierung oder Zuordnung ermöglichen. An der italienischen Neutronen-Experimentierstation INES (an der gepulsten Neutronenquelle ISIS der RAL laboratories, UK) und an der Neutronen-Computer-Tomographie- und Radiographiestation NECTAR (am FRM II-Reaktor in München) wurde eine Untersuchung der Bildgebungsmöglichkeiten an Kunstobjekten aus Metalllegierungen durchgeführt. Hierfür entstanden spezielle Proben, die in Größe und Zusammensetzung typischen antiken Kunstwerken entsprechen. Mittels Neutronentomographie konnte die innere Struktur der metallenen Testobjekte rekonstruiert und Details in ihrem Inneren auf Millimeterskala aufgelöst werden. Zudem führte man mehrere Messungen durch, um die Leistungsfähigkeit beider Anlagen gänzlich zu charakterisieren. Ihre Bildausgabe wurde hinsichtlich Linearität, Signal-zu-Rausch-Verhältnis, dynamischen Bereichs und Ortsauflösung untersucht, indem man Leuchtschirme (Szintillatoren) verschiedener chemischer Zusammensetzung und Dicke verwendete. Der Beitrag stellt eine detaillierte Beschreibung dieser Untersuchungen vor.

Keywords

neutron imaging / tomography / linearity / dynamic range / resolution