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Neutron tomography for archaeological investigations

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Abstract:
Within the last decade neutron tomography and radiography significantly gained importance. Especially its application in non-destructive testing for industrial components can be underlined. A good example is the automotive and aviation industry, where a high contrast for the used lubricants and adhesive materials is required. In contrast to X-rays, neutrons are able to penetrate thick layers of metals and provide on the other hand a high sensitivity to hydrogen containing materials. In recent years a large number of applications in other fields like biology, medicine, geology and especially archaeology have been reported. Here the potential of neutron tomography for investigations on archaeological samples shall be lined out and some recent examples will be presented.

1. Introduction

Neutron tomography has recently found new applications in many different fields like for example in biology, medicine, geology, archeology and heritage conservation [1,2,3,4]. One of the reasons is the fast development in digital image recording and processing, which enables the computation of tomographic reconstructions from high-resolution images on a reasonable timescale. The development of new detectors with better signal-to-noise characteristics and faster read-out electronics allowed to overcome some of the limits of conventional neutron radiography and tomography concerning spatial and time resolution [5].
First results applying neutron tomography for non-destructive investigation on archeological samples were reported by B. Schillinger at the 5th World Conference on Neutron Radiography held in Berlin in 1996 [4]. In the following years a growing number of tomographic measurements with neutrons have been performed on such kinds of samples [6,7]. The aim was always to obtain additional and - compared to other methods - complementary information on valuable archeological and historical objects without destruction or damage [8].
The ability of neutron radiation to penetrate thick layers of metals and to detect small amounts of hydrogenous materials makes neutron tomography a unique tool to investigate metal samples like historical weapons or jewelry and to characterize the structure of fossils and stone samples as well.

2. Neutron Radiography

2.1.Beam attenuation

The attenuation properties of materials are dependent on the specific interaction processes with the used radiation. Absorption and scattering are the interactions that contribute to beam attenuation. In conventional radiography the attenuation of the incident beam by a sample can be described by an exponential function of two parameters – the transmitted thickness

$d = \int_{path} dz$ and the distribution of the linear attenuation coefficient $\mu(x,y,z)$ in the sample (Eq. (1)).

$$I(x,y) = I_0(x,y)\exp\left[-\int_{path}\mu(x,y,z)dz\right] \qquad (1)$$

$I_0(x,y)$ and $I(x,y)$ are the intensities of the incident and transmitted beam in a plane (x,y) transversal to the propagation direction z. For high resolution radiography purposes two kinds of radiation can be used: X-rays and neutrons. The charge-free neutron interacts with the core of the atom, while in contrast X-rays interact with the charge distribution of the electron shell. Therefore the X-ray attenuation coefficients increase with the atomic number of the elements, i.e. with the number of electrons. The interaction probability of neutrons with the atomic core depends on the coherent scattering length a_{coh}, which does not show a systematic dependence from the atomic number of the element. As a consequence, the attenuation properties of the elements for neutrons show a non systematic dependence from the atomic number as shown in **Fig. 1**.

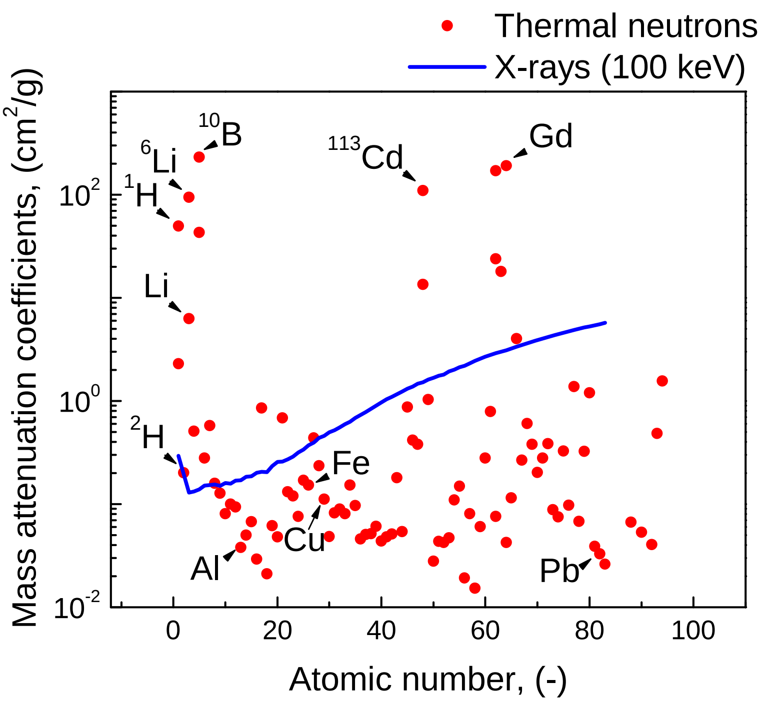

Fig. 1. Comparison of the mass attenuation coefficients for thermal neutrons and X-rays (100 keV) in dependence on the atomic number[9].

The attenuation properties besides being dependent on the energy of the applied radiation, are different for different isotopes. For radiography purposes the energy of the neutrons is typically of the order of meV (thermal or cold neutrons). X-ray energies are normally in the order of ten to several hundred keV. Comparing the mass attenuation coefficients for different elements for the case of X-rays and thermal neutrons (see **Fig. 1**) the following main conclusions can be made:

- neutrons are very sensitive to some light elements like H, Li, B, where X-rays do not provide a good contrast (low and similar atomic numbers),
- the distribution of attenuation coefficients for neutrons is independent of the atomic number which helps to achieve contrast even for neighboring elements, for X-rays one finds an approximately exponential increase with the atomic number,
- neutrons easily penetrate thick layers of metals like Pb, Fe and Cu where standard X-ray imaging facilities with energies of several hundred keV fail,
- neutrons can distinguish between isotopes (for instance ^1H and ^2H) which is not the case for X-rays.

The linear attenuation coefficients of some metals, which are of high relevance for archaeological investigations are listed in **Table 1**.

Table 1 Linear attenuation coefficients of some metals for thermal neutrons

Material	Au	Ag	Cu	Sn	Bronze	Pb	Zn	Fe
μ, [cm^{-1}]	6.28	3.99	0.99	0.20	0.87	0.37	0.34	1.20

In case of tomography investigation the sample is rotated around a defined axis where 2-dimensional projections are recorded under different rotation angles. The mathematical reconstruction of the matrix of the attenuation coefficients in the sample volume can be done using the collected set of projections.

2.2 Neutron sources

To extract an intense neutron beam for radiography purposes a large scale facility like a research reactor or a spallation source is needed. In research reactors neutrons are produced by fission of Uranium. The energy of the neutrons as produced is in the order of a few MeV. These high energies are not appropriate for conventional experimental purposes. Therefore, neutrons are moderated to energies of a few meV in a moderator, usually water or heavy water. The advantage of a steady source like a fission reactor for neutron imaging is the stable and continuous neutron flux which enables tomographic measurements with extended exposure times.
The spallation sources are accelerator-driven sources where the neutrons are produced by the excitation of the nuclei in a heavy-metal target (often Ta or W alloy). Again the fast neutrons are slowed down by collisions in a moderator set around the target. The accelerator has a defined repetition rate giving a corresponding time structure of the pulsed neutron beam. This time structure is very appropriate for time-of-flight experiments and energy-selective radiography investigations, respectively.
For high resolution neutron radiography and tomography both sources (reactors and spallation sources) provide sufficiently high neutron fluxes of app. 10^6 to 10^7 neutrons / cm^2 s at the sample positions. Typical exposure times are in the range of a few seconds, with the exception of high flux facilities (~ 10^9 neutrons/cm^{2}s) where radiographic images can be taken in fractions of a second.
An important characteristic of a neutron source is the mean energy of the neutron beam provided for experiments. As mentioned above the neutrons are moderated to thermal energies, corresponding to a few meV. Additional moderation by a “cold source”, i.e. a moderator at very low temperature (e.g. Deuterium at 30° K) is used to obtain cold neutrons. Due to their

low energy the attenuation coefficients of penetrated materials are higher for cold neutrons [10].

2.3 Beam geometry

The collimation system installed between the source and the sample is of fundamental importance for the imaging quality that can be achieved with a specific experimental set-up. The collimation system consist of several apertures manufactured from high absorbing materials like B_4C, Li or Cd which are embedded in an evacuated beam tube. The apertures are increasing along the beam propagation direction and are hence defining the beam path. However, the collimation is defined by the size of the first aperture *D* and the length of the beam path L from the first aperture to the sample position. The ratio *L/D* [11] is used to characterize the collimation of a radiographic set-up as it is limiting the spatial radiographic image resolution which can be achieved in the experiment. The higher the *L*/*D* ratio the better is the beam collimation. Typical *L*/*D* values for standard neutron radiography facilities are between 200 and 500. At a defined *L*/*D* ratio each point of the sample will be projected on the detector plane as a spot with a diameter $d=l/(L/D)$, where *l* is the distance between the sample and the detector (see **Fig. 2**). For example in the case of an *L*/*D* ratio of 500 and a given sample-to-detector distance *l* of 5 cm the corresponding spatial resolution (blur *d*) will be in the order of 100 µm. While for radiography contact images are possible, for tomographic measurements it has to be taken into account that the minimum distance *l* between sample and detector is determined by the largest dimension of the sample perpendicular to the rotation axis.

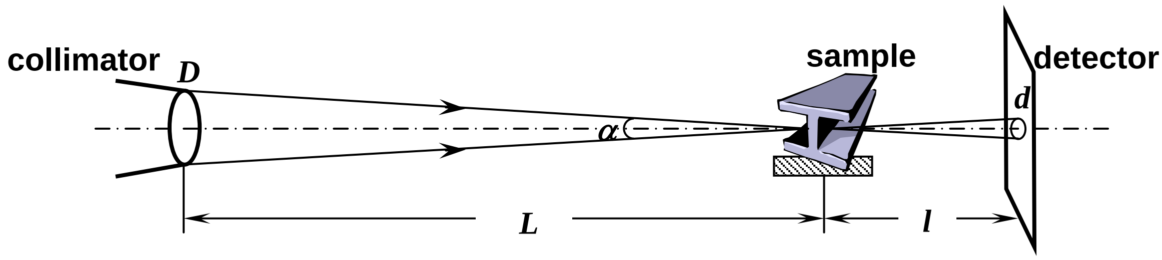

Fig. 2. Schematic representation of a neutron radiographic experimental geometry

2.4 Detectors

Other necessary components of a radiography facility are the sample manipulation system to scan - and for tomographic purposes - to rotate the sample and the 2D image detection system. Of course the achievable image resolution is very much dependent on the detector system as well. Position-sensitive detectors for high resolution imaging are based on the detection of light photons emitted from thin scintillation screens. However, neutrons cannot be detected directly by a scintillation process. Therefore an additional converter material is needed which captures the neutrons and emits some ionizing (secondary) radiation like X-rays or α-particles. Two types of converter materials are mainly used in neutron radiography detectors: ^{6}Li converting by an (n,α) and Gd by an (n,γ) reaction. This secondary radiation is then converted to light by a standard scintillating material such as ZnS. The mean free path of the secondary radiation in the scintillation material is limiting the spatial resolution of the detector to about 100 µm. A typical neutron radiography detection system is the combination of a scintillator screen and a CCD camera, which is recording the scintillation light from the screen via a mirror and a lens system. Imaging plates, flat panels and classical X-ray films (covered with a proper neutron to X-ray converter) are used as an alternative to 2D detectors.

3. The neutron tomography instrument CONRAD at the HMI Berlin

All examples of neutron tomography that will be presented were performed at the neutron tomography facility CONRAD (COld Neutron RADiography) at the HMI in Berlin (DE). It is situated at the end of a curved neutron guide facing the cold source of the BER-II reactor. The neutron guide provides a very high cold neutron flux in the order of app. 3 x 10^9 n / cm^2 s directly at the end of the guide with a negligible background of γ-radiation and fast neutrons. The facility which is not completed yet is planned with two measuring positions with very different and complementary characteristics. At the first measuring position an extremely high-flux (app. 3 x 10^9 n / cm^2 s) is available. However, on the other hand the image resolution is limited by the energy-dependent beam divergence of the guide providing a poor L/D ratio of approximately 70 [11] (resulting in a geometrical blur d = 250 µm for a distance l = 2 cm from sample to detector). Additionally the available beam size at that position is limited to a rectangular cross section of 3 x 5 cm^2. This position has been used already for first measurements some of which are presented here.

At the second position an additional collimation system, consisting of a flight tube of 5 m length and a set of apertures, will be installed in order to achieve high resolution. This way the image resolution will be improved to a value of 100 µm (L/D ~ 500). The neutron flux will be reduced but still reaching a value of app. 10^7 n / cm^2 s at the measuring position. The beam at the sample position will have a circular cross section with a diameter of 15 cm. The construction of this second position has recently started.

The detector system presently used is based on a 12-bit interline-transfer CCD camera (Sensicam) with 1280x1024 pixels. The images obtained from the LiZnS scintillator are projected via a mirror and a lens system onto the CCD chip.

4. Recent neutron tomographies of archaeological samples

4.1 Heterogeneity of archeological glass samples

Three fragments of glass from approximately the I-II century A.D. stemming from excavations in Altino near Venice, Italy [12] have been investigated at the high flux measuring position of the neutron tomography facility CONRAD at HMI. The Altino archaeological site has a complex stratigraphy, where at least three different historical layers are present, corresponding to ages starting from approximately the VII century B.C.. There is still a large amount of different items which have been recently collected there. In many cases these items have to be handled carefully and destructive analyses or analyses introducing a risk to damage an object are a problematic choice. Even cleaning the samples from clay or earth is a delicate problem. Therefore neutron and X-ray tomography are reasonable techniques to be applied in order to visualize the samples, their inner structure or for example, to look for inscriptions or engravings. Also defects (mainly due to segregation induced by corrosion) and the eventual presence of substructures and finer chromatic domains can be visualized. **Fig. 3** and **Fig. 4** are images obtained from the 3D reconstruction of a glass sample.

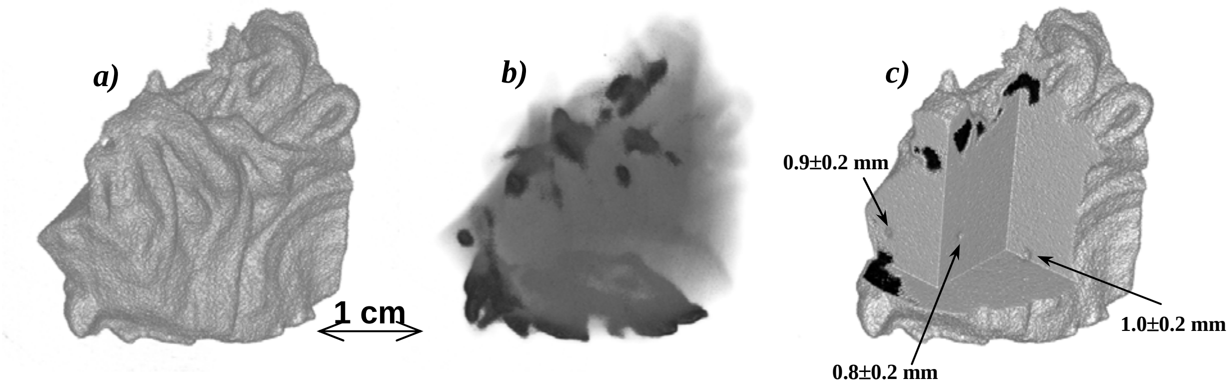

Fig. 3 Neutron tomographic reconstruction of a glass sample from the excavations in Altino. a) General view of the sample; b) high-absorbing areas; c) quantitative analysis of the voids in the glass material.

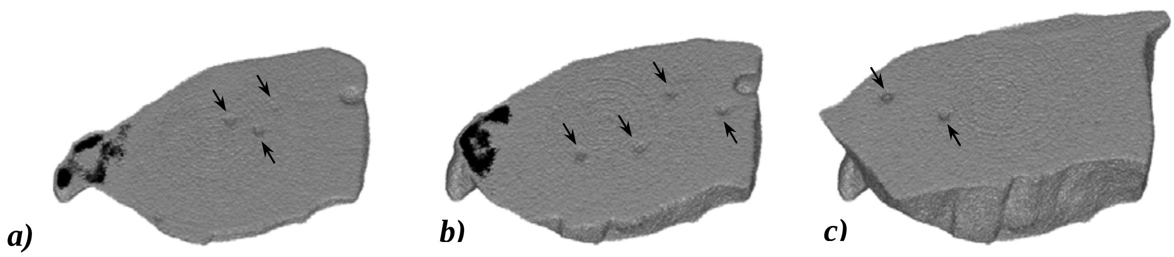

Fig. 4. Tomographic slices taken at a sample height of a) 4.1 mm, b) 6.5 mm and c) 10.7 mm

The chemical composition of the glass material has been determined by XRD. The corresponding data are presented in **Table 2**.

Table 2 The chemical composition of the glass material determined ba XRD

Material	SiO_2	Al_2O_3	CaO	Na_2O	Cl	Others (< 1%)
wt. %	67.2	2.50	7.31	19.3	1.33	Bal.

In a first qualitative analysis of the neutron tomography data areas of high-attenuation could be clearly identified as can be seen in **Fig. 3 b**. These areas could be related to earth, clay and minerals containing hydrogen mainly sticking to the sample surface. Also void structures (**Fig. 3 c**) could be found in the inner region of the sample and their sizes could be determined quantitatively. The voids can also be seen on three tomographic images representing slices at different sample heights, see **Fig. 4**. The presence of soil in inclusions and on the surface are seen as dark areas in the images. For archeology the information about the size and the distribution of the voids (marked by arrows) in the glass can be a hint concerning the glass production process in the antique.

4.2 Investigation of marble samples

Among the natural composites, there is a wide variety of stones used in buildings, monuments, statues and other objects of archaeological or cultural heritage interest. Sandstone, Limestone, Granite und Marbles are the most common stones used for these purposes. Marbles may look very different as far as texture, color, mechanical and physical properties are concerned. This is mainly a consequence of their "formation history". Here it is of archeological interest to link certain types of marbles to the areas and formations they have been taken from. Neutron tomography can provide information about the inner composition which can be used to define a kind of "fingerprint" of different types of white and polychromatic marbles in order to easily identifying their origin. Two different sets of samples were investigated. One from the island of Naxos (Greece) which is a well known source of white marbles and another one from the Villa Adriana (Tivoli, Roma, Italy) where both white and polychromatic marbles are present.

The neutron tomographic reconstruction of a polychromic marble is shown in **Fig. 5 a**. The presence of high-attenuating areas can be seen in **Fig. 5 b**. They are related to Kaolinit inclusions [$Al_2Si_2O_5(OH)_4$] in the marble matrix providing a high neutron contrast due to the containment of crystal water in their chemical composition.

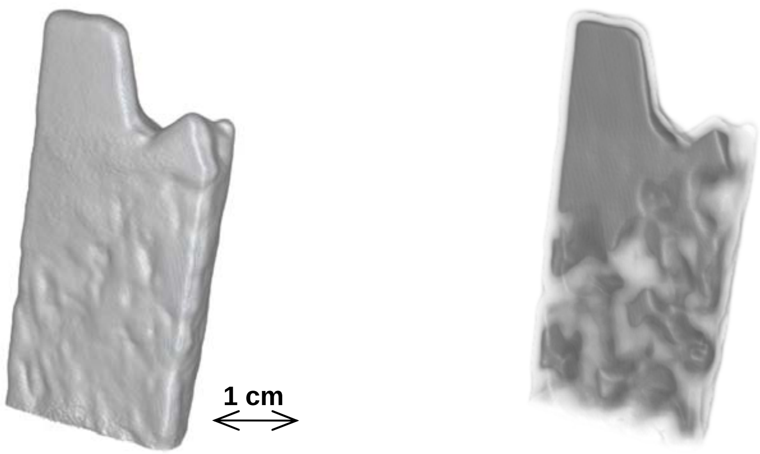

Fig. 5. Neutron tomographic reconstruction of a polychromatic marble sample from Villa Adriana. a) General view of the sample; b) Visualization of the high-attenuating areas in the sample.

The volume fraction of such inclusions in the marble matrix can be used to characterize polychromic marbles. The quantitative volume fraction (**Fig. 5**) can be determined easily from the tomographic data even by using the standard segmentation tool of the 3D-rendering software VG StudioMax [13]. It has been calculated to be approximately 43 % of the whole marble volume (V/V).

In the case of the white marble it could be found by the neutron tomographic investigation that the inner volume of the stone is much more homogeneous. Some minerals could be identified on the sample surface, see **Fig. 6**. Most likely these minerals, whose volume fraction was found to be 4.4 vol. %, are residues of material used to hold the marble in place.

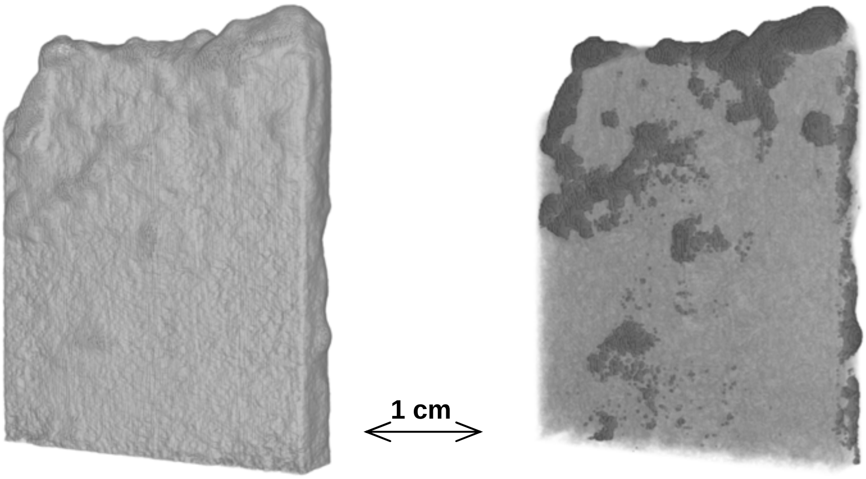

Fig. 6. Neutron tomographic reconstruction of a white marble sample from Villa Adriana. a) General view of the sample; b) Visualization of the high-attenuating areas in the sample.

Conclusions

Neutron tomography can be a powerful tool for non-destructive investigations of the inner structure of a broad range of samples. The examples presented here, like others reported before demonstrate the advantages of applying this technique to archaeological samples where it is of particular interest to avoid any damage of the object. The possibility of studying large samples (up to several hundred square centimetres) as well as thick samples of metals and of geological materials in reasonably short time, makes neutron tomography a very attractive method for addressing a large number of archaeological problems. Of course the technique is also useful for a wider variety of industrial and practical problems. However, attention has to be paid on possible neutron activation of the samples, which should be calculated preliminary in order to prevent problems with the radiation protection norms for transportation and storage of the samples.

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