KURT OSTERLOH · SANJEEVAREDDY KOLKOORI · AMÉLIE NUSSER · ANDREAS SCHWABE
THOMAS BÜCHERL

NEUTRON COMPUTED TOMOGRAPHY FOR CONCEPTS IN DECONTAMINATING BIOCIDALLY TREATED WOODEN ARTEFACTS

At the beginning of the 20th century, biologically degraded wooden items in churches such as carved statues or organ lofts have been preserved with pesticides or with *carbolineum* as it was common practice with wooden railway sleepers. This treatment resulted not only in polluting the room atmosphere but also developing dark stains on the surface leaving a horrible appearance, particularly in an annoying manner on those of retables and epitaphs. As a consequence, the biocidal treatment not only raised an aesthetic problem but also an environmental one. As the dark stain on the surface appears in a spotted manner it has to be assumed that the impregnation material may also be heterogeneously distributed or migrates along certain internal structures. Therefore, any knowledge about the interior of the affected objects could provide an essential support in guiding a way for an appropriate decontamination procedure. The only way of obtaining such a view is a non-destructive inspection, the method of choice is radiography and tomography. Since both, the impregnant, i.e. the preservation agent, and the wooden matrix consists of hydrocarbons, X-rays are capable to reveal the internal structures based on the thicknesses of the penetrated layer but hardly any material differences. The tool of choice to detect contrast differences due to variations in the hydrocarbon concentrations that is expected to be increased by the impregnant content could be another kind of radiation, i.e. neutrons. They are particularly absorbed by hydrogen, i.e. hydrogen containing compounds are giving the contrast to the radiographic images (Bücherl/Lierse von Gostomski 2011; Osterloh et al. 2008). However, the penetration depth of neutrons through wood depends on their energy (Osterloh et al. 2011b). Since the interrogated specimens were larger than a fewcm in thickness, it was necessary to use a facility with fast neutrons (1.5-2 MeV) such as the NECTAR instrument at the FRM II research facility in Garching, Germany (Bücherl et al. 2011). The first part of the results is supposed to demonstrate the potential and the limits of neutron radiography and tomography with wooden specimens. The investigated objects, two sculptures taken from an epitaph in a church in Northern Germany, will be presented in the second part. Finally, the results obtained with the neutron technologies will be compared with their X-ray counterparts.

Materials and methods

The specimens, i.e. two sculptures representing an angel and a female person called the »Virtue of Hope«, were taken from the baroque epitaph Reyer (1704) at the St. Laurentius church in Tönning (Schleswig-Holstein/Germany). The impregnating agent has chemically been identified as *carbolineum*, a mixture of coal tar oil components containing polycyclic aromatic hydrocarbons (PAH), predominantly anthracene, phenanthrene, fluoranthene and pyrene as found in samples collected from the surface. The polycyclic aromatic hydrocarbons are classified as carcinogenic and harmful to the environment. All specimens were stored in a climate controlled room at the Rathgen Research Laboratory.

Neutron radiography and computed tomography (CT)

The NECTAR facility at the Forschungs-Neutronenquelle Heinz Maier-Leibnitz (FRM II) in Garching, Germany, was chosen for this study since it is one of the very few sites where fast neutrons are available which were needed for this purpose (Bücherl et al. 2011; Osterloh et al. 2011b). At this site, a continuous stream of fission neutrons is generated in converter plates in marginal position within the moderator tank of the reactor showing a typical fission spectrum with its maximum about 1.8 MeV. The beam is conducted through a set of filters mainly to reduce gamma radiation and thermal neutrons. The length per diameter (L/D)-ratio in this setting was 230. The detection system consisted of a fluoroscope with a pp-converter plate with ZnS(Ag) as a scintillator and an ANDOR DV434-BV CCD camera. Generally, the exposure time was 60 s per single image, five images were taken per projection. The object-detector distance was 40 cm. Subsequently, the image processing consisted of a dark image and a bright field correction, automatic removal of spotty artefacts (Osterloh et al. 2011a; 2012), median of single images (usually 5); the CT was performed in the range of 0-180° projections in 1° steps. In order to maintain the original grey values as far as possible, the Fourier slice theorem (Buzug 2008) was used for reconstruction with back-projection in the Fourier-transformed (FT) space without filtering that was found sufficiently suitable for parallel beam geometry (Osterloh et al. 2011b; 2011c; 2011d).

X-ray radiography and computed tomography (CT)

The X-ray radiography and computed tomography (CT) was carried out in the laboratories of the BAM Federal Institute for Materials Research and Testing, Division of Radiology, in Berlin. A Comet X-ray tube type Y.TU 600-D01 served as the radiation source. The settings were 160 kV and 4 mA. The radiographic images as well as the CT projections were recorded with a Perkin- Elmer flat panel detector, type XRD 1621AN16 with 2048 × 2048 pixels of 200 × 200 µm size. The exposure time was 15 s per single frame, the source-detector distance 2342 mm and the object-source distance 1692 mm. The CT consisted of 1000 projections in the range of 0-360°. The reconstruction was achieved with the Volume Graphics software VGStudio MAX 2.2 and its supplement for tomography.

Results

First, the potentials and the limits of radiography and tomography using fast neutrons will be presented with wooden specimens of different sizes, from a piece of log to a larger section of glulam. Two sculptures taken from the epitaph mentioned in »materials and methods« are subjected to neutron tomography, one of them also to X-ray tomography to show the qualitative differences between these two technologies.

Radiography and tomography of large wooden specimens

The first wooden specimen (**fig. 1, 1**) interrogated with fast neutrons demonstrated the necessity of a particular image processing in order to get rid of the massive occurrence of spotted interferences impairing the visibility of the true objects (**fig. 1, 2**). For this purpose, a general algorithm has been developed to remove these stochastic artefacts while leaving all other image information untouched (Osterloh et al. 2011a; 2012). The final result was achieved by calibrating the radiograph with an open beam image (without object) treated in the same manner (**fig. 1, 3**). The second specimen in this set of experiments showed the capability of penetrating even thick layers of heavy metal, in this case a thick steel plate, while still showing the annual rings in the

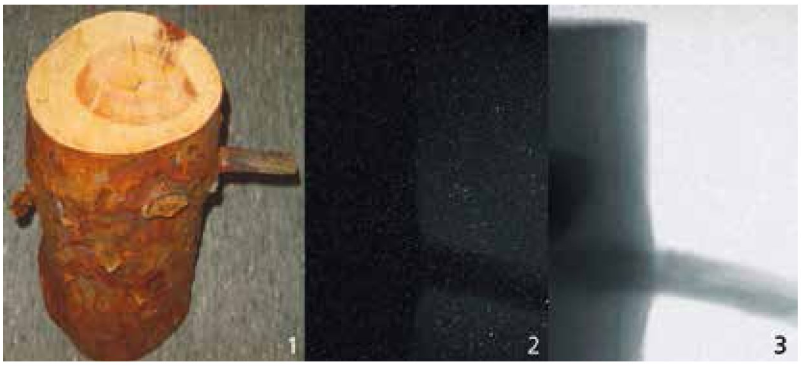

Fig. 1 Piece of log (**1**) with the unprocessed neutron radiograph (**2**) affected with spotted flaws and after appropriate image processing (**3**). – (Photo, X-ray and image processing K. Osterloh).

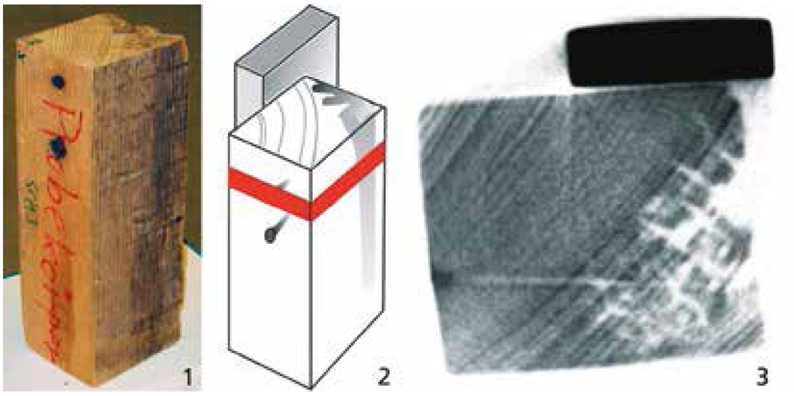

Fig. 2 **1** small wooden girder with holes filled with plasticine. – **2** sketch showing the position of a thick steel plate and of the tomographic section. – **3** neutron tomograph with a cross section as indicated. – (Photo, drawing and CT K. Osterloh).

direct vicinity of that shielding (**fig. 2, 1**). On the other hand, a hole stuffed with plasticine is clearly visible on the left side of the image (**fig. 2, 2**). Finally, the limit of penetration is reached at some 50 cm as shown with the piece of glulam (**fig. 3**), though with the aid of a filtering procedure. This processing of the radiographic image (Osterloh/Zscherpel 2012) enhancing the structural details showed the position of internal knots, the Lucite block added to the specimen and faintly some annual rings. It was possible to show some internal details of this specimen by tomography as it was also possible with smaller specimens not exceeding 2 cm of layer thickness applying thermal neutrons (Osterloh et al. 2011b; 2011c; 2011d). Based on the parallel beam geometry, it was possible to achieve images of details in certain regions of interest even with specimens of a size partly exceeding the viewing field of the instrument, i. e. the size of the detector plane.

The sculpture »Virtue of Hope« and the angel sculpture

The sculpture illustrating a woman is named »Tugend der Hoffnung« that translates into »Virtue of Hope«. The neutron radiographs (**fig. 4**) in frontal (**fig. 4, 2**) and lateral direction (**fig. 4, 3**) were not very conclusive for evaluating density distributions because various distances have been passed by the neutrons through the specimen. As a consequence, it was necessary to proceed with the tomographic approach. The tomographic reconstruction purposely omitted any filtering procedure to maintain the voxel intensities as accurate as possible since the purpose of this technology was to obtain density information rather than structural details. The appropriate technology to reveal the latter ones much more precisely was X-ray tomography (see below). The cross section obtained from the reconstructed data set (**fig. 5**)

Fig. 3 **1** penetration of fast neutrons through a layer of 50 cm wood. – **2** beam direction from left to right, radiographic image showing the shade of the Perplex block at the bottom (red arrows), filtered radiographic image showing several internal knots more clearly. – (Photo and processed neutron radiographic image K. Osterloh).

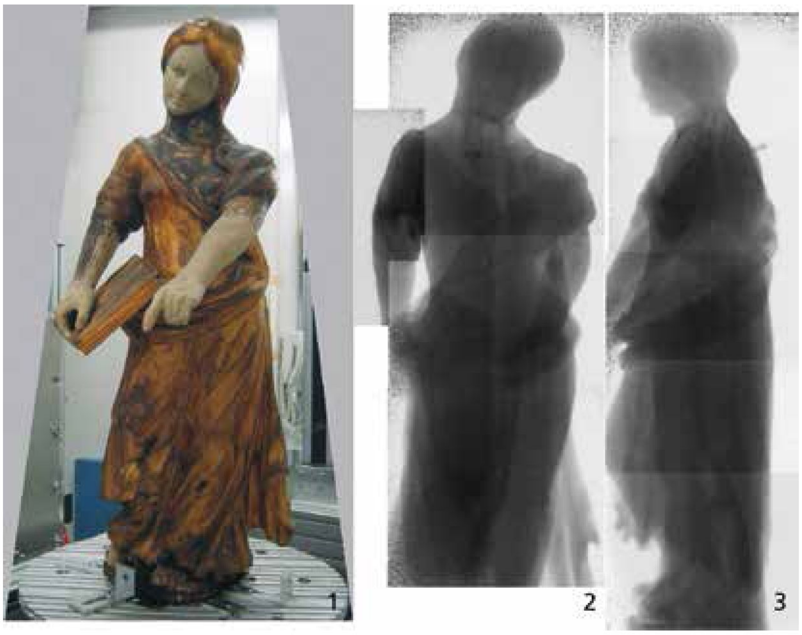

Fig. 4 Sculpture »Virtue of Hope« (**1**) with neutron radiographs in frontal (view from behind) (**2**) and lateral direction of penetration (**3**). – (Photo and composed neutron radiographic image by the authors).

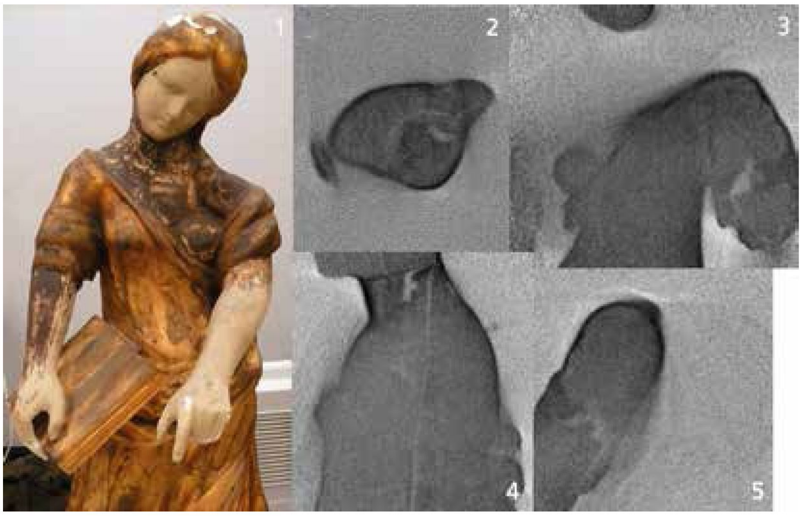

Fig. 5 Neutron CT of the sculpture »Virtue of Hope« (**1**) with a cross section in the lower neck region (**2**) and longitudinal sections in different directions (**3-4**). The region in the upper left arm (**5**) shows indications of repair (frontal in the upper right panel and saggital in the lower right one) matching with the relative spotless appearance in the photograph (left). – (Photo and processed neutron CT images by the authors).

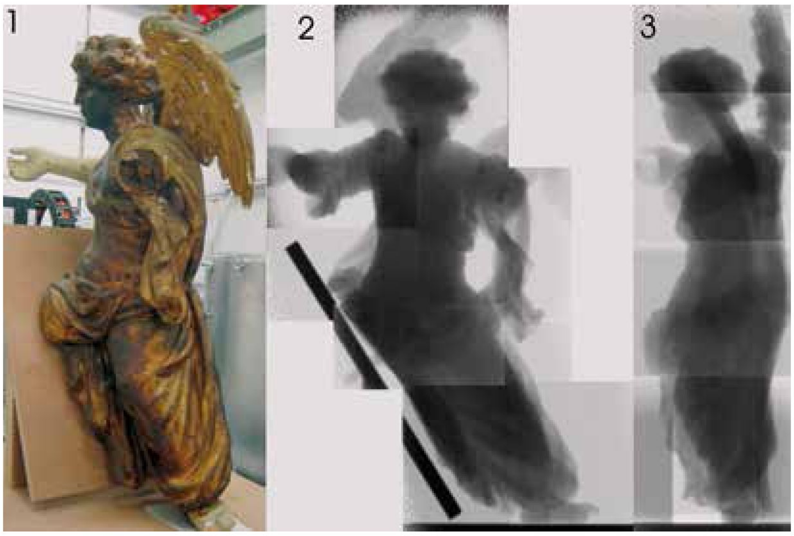

Fig. 6 Neutron radiograph of the angel sculpture in two different directions, together with the jig keeping the specimen in position. – (Photo and composed neutron radiographic image by the authors).

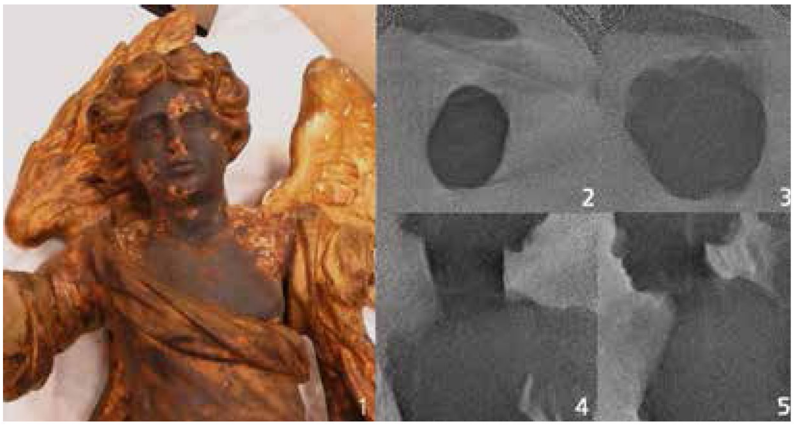

Fig. 7 Neutron CT (**1**) of the angel sculpture with cross sections from different regions (**2** neck; **3** head; **4-5** longitudinal sections in different directions). – (Photo and neutron CT images by the authors).

showed a rather heterogeneous interior with an outer shell of higher density. Furthermore, there is a dense layer between the body and the head which was obviously replaced by a restoration measure in the past (fig. 5, 4-5).

The same problem of evaluating densities in a radiographic image was encountered with the other sculpture illustrating an angel (fig. 6). It was necessary to apply the tomography technology for this purpose. Cross sections in the same manner as with the previous sculpture again showed a heterogeneous density distribution with a dense layer at the surface which, however, may be partly also related to the paint covering. Further chemical analysis will be necessary to elucidate whether the density might be contributed, at least in part, to a higher impregnant concentration (whatever kind it might be).

Comparison between neutron and X-ray radiography tomography

The first sculpture »Virtue of Hope« was also subjected to X-ray radiography and tomography (fig. 8, 2. 4. 6). The radiographic images revealed some nails or certain plaster layers possibly containing calcium for reinforcement within the body of the sculpture from previous restoration works (fig. 8, 2, red circles) but invisible with neutrons (fig. 8, 1, red circles). Structural details enhanced by filtering (fig. 7, 2) in the way as described before. As expected, the X-ray tomography revealed much more structural details as the neutron technology but with a slightly different density distribution (fig. 8). Due to the profoundly different positioning, beam geometries and detector technologies it was impossible to

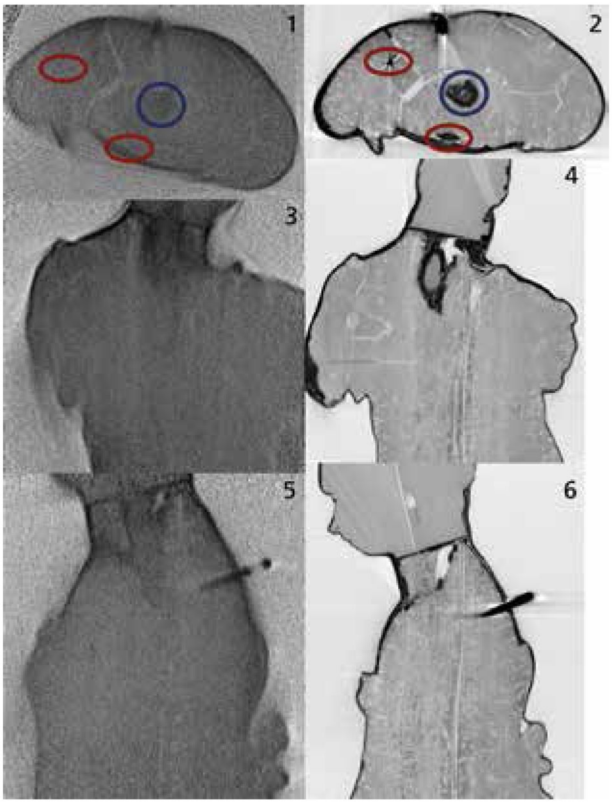

Fig. 8 Sculpture »Virtue of Hope« (see **figs 4-5**), comparison between neutron (left) and X-ray (right) tomographs: **1-2** cross section. – **3-4** longitudinal section with frontal view. – **5-6** lateral section with sagital view. The features in the centre of the cross section (blue circle) show different intensities with neutrons and X-rays. Nails or plaster layers (calcium?) are only visible with X-rays (red circles). – (Neutron CT images by the authors, X-ray CT with the assistance of C. Bellon and S. Hohendorf, whose help is gratefully acknowledged).

match the sections after the reconstructions from both data sets. Nevertheless, sections from the same regions within the sculpture could be compared. For this purpose, the screw with the eye on the back of the sculpture served as a point of orientation. Interestingly, the lumen of the eye was visible with neutrons (**fig. 8, 5**) but not with X-rays (**fig. 8, 6**). This can be explained by the different penetration abilities of neutrons and X-rays through heavy metals. Rather obviously, a pole has been inserted in the centre in one of the previous restoration works. Furthermore, this pole appeared darker in the X-ray sections as in the neutron ones. Different contrast distributions have been detected in the neck region. The contact layer between the body and the head was clearly visible with both technologies. The X-ray images clearly showed that wood in the head region was less biologically damaged and that the head and the body consist of different types of wood. These results indicate that the head was renewed during the restoration. The cleft running through the head was also visible at the outside where the paint cover was chipped off. Maybe that the matrix of the head was less soaked with the *carbolineum* leaving the face of the sculpture appearing rather faultlessly as compared to the other parts of the outer surface.

Summary, conclusion and outlook

Large wooden objects of cultural heritage that have been impregnated with *carbolineum* were successfully interrogated with fast neutrons to study the internal density distribution that may give a clue to the location of the impregnating agent in context of the internal structure since the dark flaws on the surface appear heterogeneously. Some peculiar problems of neutron radiography with large wooden specimens have successfully been resolved. It became obvious that computed tomography (CT) was essential to study density distributions in such objects of an irregular shape where penetrating radiation passed different layers of thicknesses. In this study, the available CT technologies with X-rays and neutrons were successfully applied for Non-Destructive Evaluation (NDE) of cultural artefacts. The purpose of neutron CT was predominantly to study density distributions leaving the detection of structural details to the X-ray CT. The endogenous heterogeneity has been revealed by both technologies so both of them form the base for further studies, particularly for determining the sites of sample taking for further chemical analysis. It is intended to continue this kind of studies along the course of a selection of decontamination procedures. The knowledge of internal structural details will be also important to assess the mechanical strength of certain parts of the investigated objects.

Acknowledgements

This project was supported by the Deutsche Bundesstiftung Umwelt (DBU) under the grant Az. 30165 – 45. The valuable help received from several colleagues is gratefully acknowledged, namely from B. Redmer, S. Hohendorf and C. Bellon for achieving the X-ray tomography results and from M. Freitag, N. Wrobel, S. Simon and U. Ewert for many fruitful discussions.

References

Bücherl / Lierse von Gostomski 2011: Th. Bücherl / Ch. Lierse von Gostomski, Real-time radiography at the NECTAR facility. Nuclear Instruments and Methods in Physics Research A 651 (1) 2011, 175-179. DOI:10.1016/j.nima.2011.01.061.

Bücherl et al. 2011: Th. Bücherl / Ch. Lierse von Gostomski / H. Breitkreutz / M. Jungwirth / F. M. Wagner, NECTAR – A fission neutron radiography and tomography facility. Nuclear Instruments and Methods in Physics Research A 651 (1) 2011, 86-89. DOI:10.1016/j.nima.2011.01.058.

Buzug 2008: T. M. Buzug, Computed tomography, from Photon Statistics to modern Cone-Beam CT (Berlin, Heidelberg 2008) 163-167. DOI:10.1007/978-3-540-39408-2.

Osterloh / Zscherpel 2012: K. Osterloh / U. Zscherpel, Ways of digital image processing to obtain fast and unbiased Results. 18th World conference on nondestructive testing, 16-20 April 2012, Durban / South Africa. www.ndt.net/article/wcndt2012/papers/284_wcndtfinal00284.pdf

Osterloh et al. 2008: K. Osterloh / Ch. Raedel / U. Zscherpel / D. Meinel / U. Ewert / Th. Bücherl / A. Hasenstab, Fast neutron radiography and tomography of wood. Insight 50 (6) 2008, 307-311.

Osterloh et al. 2011a: K. Osterloh / Th. Bücherl / Ch. Lierse von Gostomski / U. Zscherpel / U. Ewert / S. Bock, Filtering algorithm for dotted interferences. Nuclear Instruments and Methods in Physics Research A 651 (1) 2011, 171-174. http://dx.doi.org/10.1016/j.nima.2011.01.107.

Osterloh et al. 2011b: K. Osterloh / D. Fratzscher / M. Jechow / Th. Bücherl / B. Schillinger / A. Hasenstab / U. Zscherpel / U. Ewert, Limited view tomography of wood with fast and thermal neutrons. International Symposium on Digital Industrial Radiology and Computed Tomography 2011, Poster 14 BB 128 CD. www.ndt.net/article/dir2011/papers/p14.pdf

Osterloh et al. 2011c: K. Osterloh / D. Fratzscher / A. Schwabe / B. Schillinger / U. Zscherpel / U. Ewert, Radiography and partial tomography of wood with thermal neutrons. Nuclear Instruments and Methods in Physics Research A 651 (1) 2011, 236-239. DOI:10.1016/j.nima.2011.02.095.

Osterloh et al. 2011d: K. Osterloh / D. Fratzscher / U. Zscherpel / U. Ewert / A. Hasenstab / Th. Bücherl / B. Schillinger, Partielle Tomographie mit Neutronen zur Beurteilung von Leimschichten in Holz. Jahrestagung der Deutschen Gesellschaft für zerstörungsfreie Prüfung (DGZfP) 2011 - Mi.4.B.2, BB 127 CD. www.ndt.net/article/dgzfp2011/papers/mi4b2.pdf

Osterloh et al. 2012: K. Osterloh / Th. Bücherl / U. Zscherpel / U. Ewert, Image recovery by removing stochastic artefacts identified as local asymmetries. Journal of Instrumentation 7, 2012, 1-14. C04018 DOI:10.1088/1748-0221/7/04/C04018; http://iopscience.iop.org/1748-0221/7/04/C04018

Summary / Zusammenfassung

Neutron Computed Tomography for Concepts in Decontaminating Biocidally Treated Wooden Artefacts

The preservation of wooden artwork using pesticides or *carbolineum* some hundred years ago raised new problems over the years. They consist not only of aesthetic ones such as darkening of pigmentations but also of environmental ones by the persisting release of polluting substances. Approaches of resolving these problems should be preceded by a vision into the inside situation of the infested objects. Radiological methods with fast neutrons were chosen as appropriate measures for this purpose since imaging with neutrons is sensitive to reveal density distribution patterns of hydrocarbons inside of an object. It was expected that the treatment with *carbolineum* had

Neutronen-Computer-Tomographie für Konzepte zur Dekontamination von mit Bioziden behandelten hölzernen Artefakten

Die vor einigen Hundert Jahren vorgenommene Konservierung von Kunstwerken aus Holz mit Pestiziden oder *Carbolineum* hat über die Zeit neue Probleme verursacht. Diese sind nicht nur ästhetischer Natur wie das Nachdunkeln von Pigmenten, sondern belasten durch die ständige Freisetzung von giftigen Substanzen auch die Umwelt. Bevor man an die Lösung dieser Probleme herangeht, sollte ein Blick auf die Situation im Inneren der verseuchten Objekte erfolgen. Als dazu geeignete Maßnahmen wurden radiologische Methoden mit schnellen Neutronen gewählt, da die Bildgebung mit Neutronen sensibel auf das Dichteverteilungsmuster von Kohlenincreased the hydrogen content of the wooden matrix. Fast neutrons were demonstrably essential to penetrate the interrogated specimens. Imaging problems encountered with fast neutrons have been successfully resolved, metal shielding did not impair the detection of structural details, and the ultimate penetration limit in wood has been reached at a layer thickness of 50 cm. Two sculptures taken from an epitaph in a church in Northern Germany were studied in detail, one of them additionally with X-rays. It was shown that computed tomography (CT) was much more conclusive than straight forward radiography. While X-ray techniques remained the measures of choice to investigate structural details neutron CT reflected the hydrogen content that may give a clue to the distribution of the impregnant the objects were treated with. It is suggested to use a combination of both technologies to support the preservation work on wooden artworks infested with the described problem.

wasserstoffen im Inneren eines Objekts reagiert. Man erwartete, dass die Behandlung mit *Carbolineum* den Wasserstoffgehalt der hölzernen Matrix erhöht hat. Schnelle Neutronen waren nachgewiesenermaßen erforderlich, um die untersuchten Proben zu durchdringen. Probleme bei der Bildgebung mit schnellen Neutronen ließen sich erfolgreich lösen, die metallene Abschirmung behinderte nicht den Nachweis von strukturellen Details, und die endgültige Grenze für die Durchdringung von Holz wurde bei einer Lagendicke von 50 cm erreicht. Zwei Skulpturen von einem Epitaph einer Kirche in Norddeutschland wurden im Detail untersucht, eine von ihnen zusätzlich mit Röntgenstrahlen. Es ließ sich zeigen, dass die Computer-Tomographie (CT) viel schlüssiger war als nur einfache Radiographie. Während Röntgentechniken erste Wahl blieben, um strukturelle Details zu untersuchen, gab die Neutronen-CT den Wasserstoffgehalt wieder, der einen Hinweis auf die Verteilung des Mittels geben kann, mit dem die Objekte imprägniert wurden. Es wird vorgeschlagen, an hölzernen Artefakten, die von dem beschriebenen Problem betroffen sind, eine Kombination beider Techniken anzuwenden, um die Konservierungsarbeiten zu unterstützen.

Keywords

computed tomography / radiology with fast neutrons / interrogation of wooden specimens with neutrons / *carbolineum* / preservation of wooden artefacts