

NEUTRON TOMOGRAPHY OF ARCHAEOLOGICAL WATERLOGGED WOOD

As an organic material, wood is prone to decay. However, under specific conditions, even very old wooden artefacts can be found in apparent good state. That is the case for wood found in wet places, where it has been waterlogged during its burial time (Hoffmann/Jones 1989). Nevertheless, archaeological waterlogged wood underwent changes in chemical composition and mechanical strength that make it very different from recent wood. Through hydrolysis or fungi attack, holocellulose in the cell walls decomposes, leaving only a lignin network to support the wood. Even the lignin will break down over a long period of time. These changes make the wood more porous and permeable to water (Unger/Schniewind/Unger 2001). The shape of the wood is preserved by the remaining lignin structure and the absorbed water, as long as the wood is kept wet. But when it is exposed to ambient conditions, the drying often leads to drastic shrinkage and collapse of the object. This collapse is believed to occur due to the loss of strength of the cell walls that cannot withstand anymore the drying stresses generated by the surface tension of liquid water in air (Grattan/Clarke 1987; Hoffmann/Jones 1989).

A way to avoid the drying stresses is to remove the water by sublimation, by freeze-drying the wood. However, it requires freezing the water beforehand and thus introduces new potential damage due to the crystallization of ice.

Freeze-drying is thus often used in conjunction with a cryoprotectant. Poly-ethyleneglycol (PEG), available in different molecular weights, is widely employed in this respect (Kaye/Cole-Hamilton/Morphet 2000). It has also the advantages of alleviating the shrinkage by replacing the bound water, and of adding strength to the wood after drying. It is the procedure routinely used in the conservation department of the Archäo-

logische Staatssammlung München, and the reason why it is studied here.

Nevertheless, the conservation treatment is still prone to questioning. In particular, how the distribution of the polymer in the wood structure affects the effectiveness of the treatment remains unclear.

Computer tomography offers the possibility of visualizing an object in 3D from 2D projections, without physically cutting the object. Neutron tomography, despite detector systems offering relatively lower spatial resolution capabilities than the ones used for X-Ray tomography (Bugani et al. 2008), has the advantage of offering a higher sensitivity to hydrogen and is thus often used to study organic materials.

The present work investigates the possibility of using neutron tomography to visualize the distribution of PEG in archaeological waterlogged wood, after the conservation treatment.

Experimental procedure

Origin and preparation of the samples

The wood used in the study is a branch of alder found in a Roman well in East Bavaria, Germany, where it remained under anoxic conditions before its excavation and kept immersed in water during one year after. Before the conservation treatment began, it was still homogeneous without cracks. Even though it is not necessarily a sample of precious cultural heritage, it allows studying various approaches of treatment. The branch has been cut in eighteen slices with a thickness of 2 cm that have been impregnated during different durations. Three samples have been impreg-

nated during eight hours, six during one week and eight during ten weeks, and subsequently freeze-dried. The impregnation has been performed in heated containers filled with a solution of PEG 1500. The concentration of the solution was 25 % in weight for all the samples, the solution was stirred regularly and the temperature of the bath was at minimum 55°C. The freezing step has been carried out at a temperature of -40°C and freeze-dried at the same temperature but under a pressure of 2.7 mbar. Eventually, the slices have been cut again after the treatment to a dimension of 2 cm × 1.5 cm × 7 cm (thickness × width × height) to permit a sufficient transmission of the neutrons. It has to be noted here that 2.5 cm was the maximum thickness allowed by the neutron beam to obtain a radiograph.

Experimental setup at ANTARES

The tomographs have been made in 2009 at the ANTARES facility in the neutron source FRM II in Garching, Germany. A polychromatic beam and a ratio L/D of 400 have been chosen for minimizing the exposure time, giving a maximal neutron flux on the order of $10^8 \text{ cm}^{-2} \text{ s}^{-1}$ with energies between 1 and 100 meV. The energy spectrum of the cold neutrons beam used in ANTARES, in the setup of 2009, can be found in (Grünauer 2005; Schulz et al., this volume). The samples have been attached to the aluminium sample holder at a distance of 15 mm from the scintillation screen, made of Li/ZnS with a thickness of 100 µm. The field of view has been set to 10 cm × 10 cm. The chosen exposure time was twelve seconds, except for the samples impregnated during ten weeks where 15 s were necessary. It allowed a tomograph, consisting of the reconstruction of 800 radiographs, to be done in less than four hours.

Segmentation of the histograms

The segmentation of the histogram is the attribution of a specific colour to a specific range of attenuation values, to reveal the spatial distribution of a material

or a mixture of materials. In our experiment, the attenuation of a mixture of wood and PEG, \sum_{sample} , is measured:

$$\sum_{\text{sample}} = \sum_{\text{wood}} + \sum_{\text{PEG}}$$

The attenuation of air is neglected, since it is well below the attenuation of both wood and PEG. It has been chosen to stretch the segmentation from the attenuation due to a non-impregnated wood to the attenuation of the PEG alone. The attenuation coefficients \sum_{wood} and \sum_{PEG} of the pure materials have been measured by doing a tomograph of the same wood not impregnated with PEG, but freeze-dried, and a tomograph of a solid piece of pure PEG 1500. For the neutron energy spectrum used in ANTARES, they have been found to be $\sum_{\text{wood}} = 414$ and $\sum_{\text{PEG}} = 3868$ (in arbitrary units), respectively. Let us call x the contribution of the PEG to the attenuation of one voxel. Here we assume that the attenuation of a mixture of wood and PEG cannot exceed the attenuation of the pure PEG. Consequently, when $x = 1$, the attenuation is only due to the presence of solid PEG. When $x = 0$, the attenuation is only due to a wood which has not been impregnated with PEG. The previous expression can thus be written as follows:

$$\sum_{\text{sample}} = x \cdot \sum_{\text{PEG}} + (1 - x) \cdot \sum_{\text{wood}}$$

This equation is transformed to:

$$\sum_{\text{sample}} = x \cdot (\sum_{\text{PEG}} - \sum_{\text{wood}}) + \sum_{\text{wood}}$$

By replacing by the values of attenuation, it comes:

$$\sum_{\text{sample}} = 3454 x + 414$$

The segmentation has been done by range of 15 % of the attenuation, and artificial colours have been attributed to every range to allow an easier visualization. Figure 3 shows three samples that have been segmented in that way.

Results and discussion

A tomograph allows a direct visualization of an object in its three dimensions, but also a selection

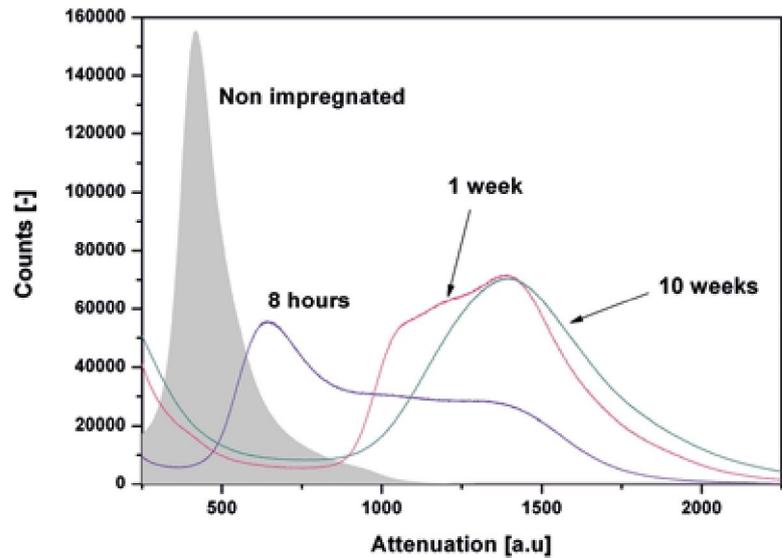


Fig. 1 Examples of histograms of the non-impregnated wood and the wood impregnated during eight hours, one week and ten weeks. – (Th. Demoulin).

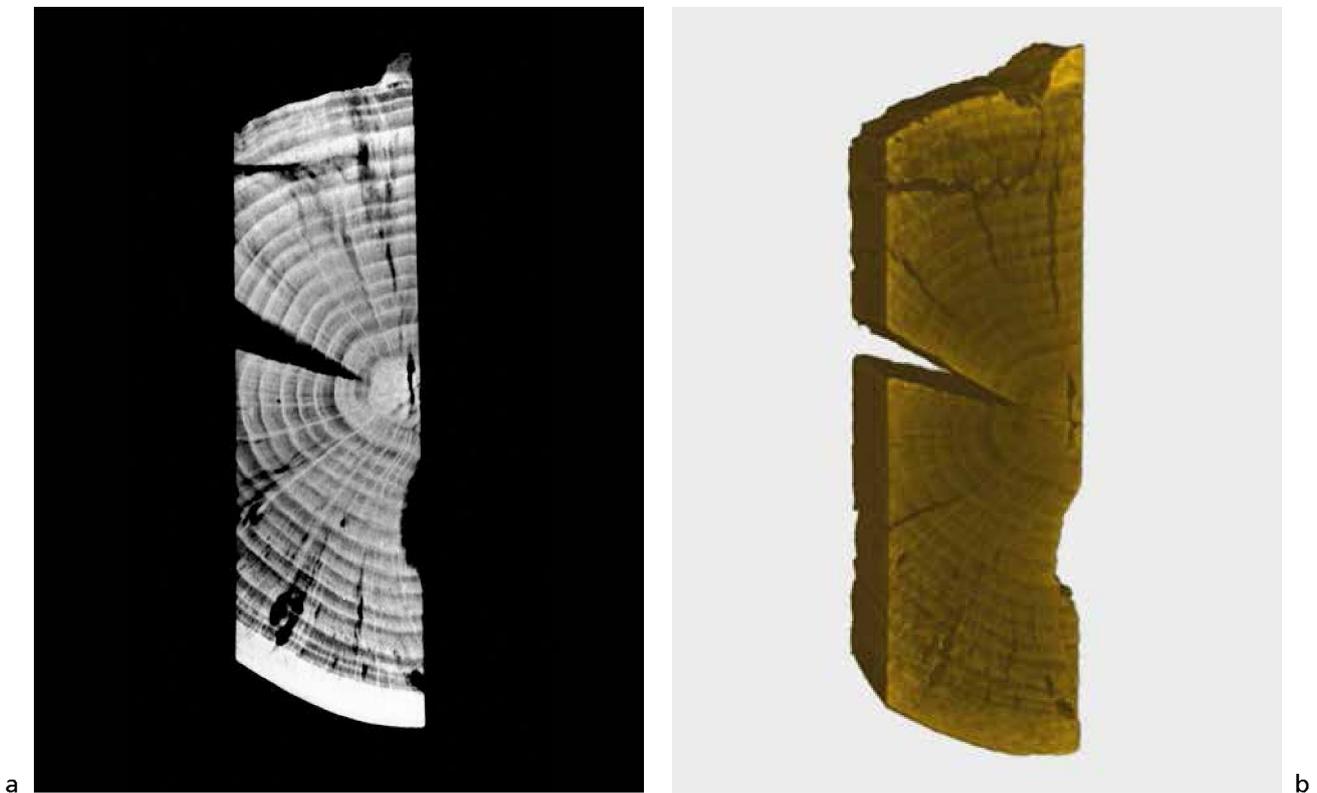


Fig. 2 **a** neutron radiograph (in inverted colour). – **b** tomograph of a sample of alder wood. – (Th. Demoulin).

and quantification of a volume inside the object. The selection is made possible through the histograms that reflect the volume occupied by a certain attenuation coefficient. The histograms for the non-impregnated wood, and for samples impregnated dur-

ing eight hours, one week and ten weeks are visible in figure 1. As a first observation, they do not allow to distinguish the PEG from the wood. The higher hydrogen density of the polymer would allow a large gap between the attenuation of the wood and the

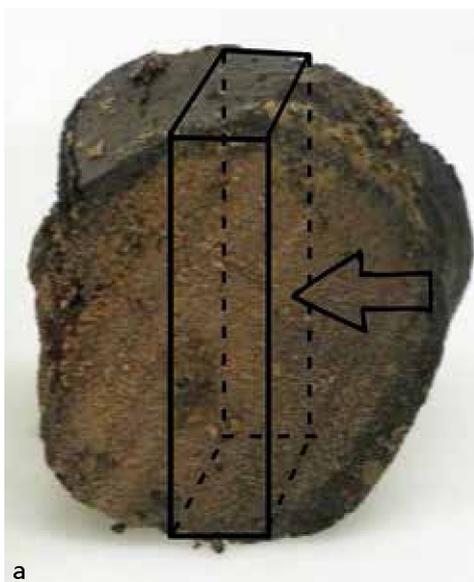
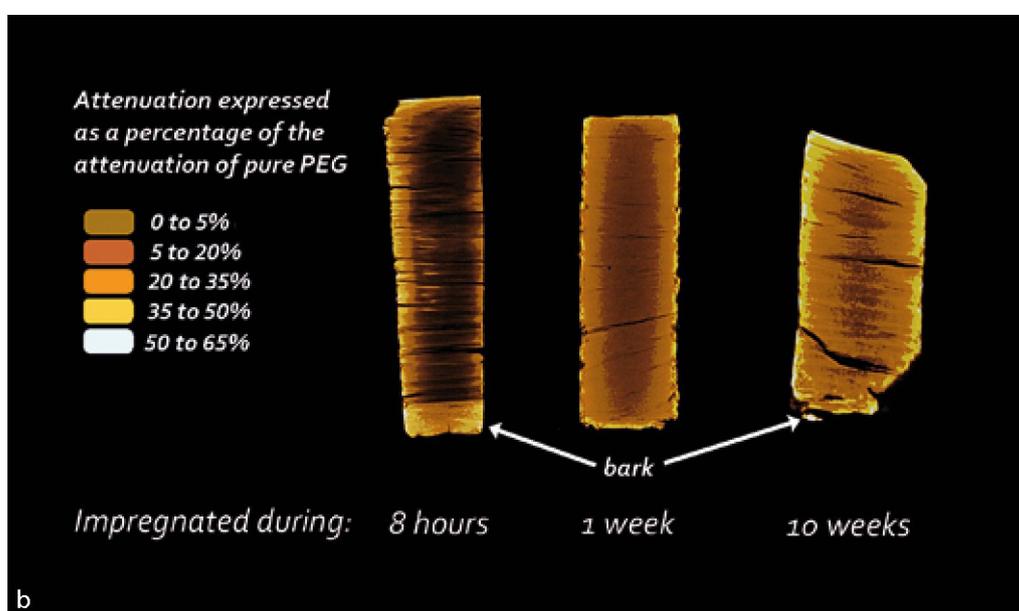


Fig. 3 **a** view of a sample before cutting. The black box symbolizes how the sample is cut and the arrow the direction of visualization in the tomograph. – **b** visualization of the attenuation in samples having different duration of impregnation. – (Th. Demoulin).



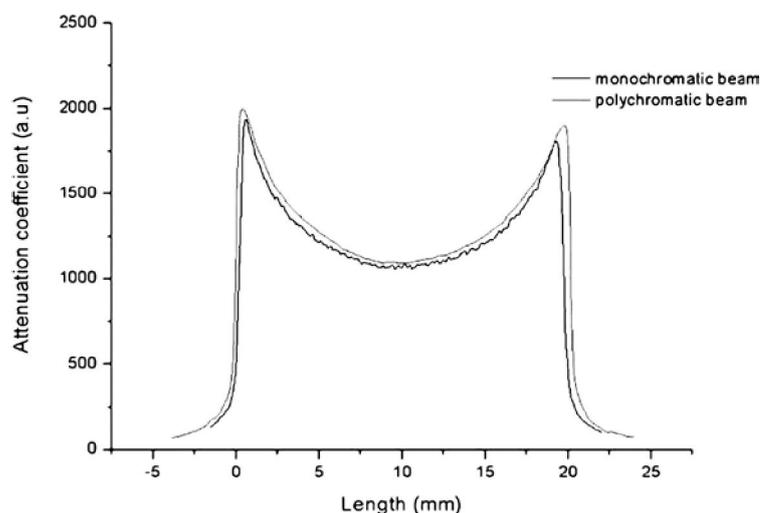
PEG, but the concurrent effect of a relatively low spatial resolution does not make a discrimination possible. A single radiograph, as shown in figure 2, had a maximum resolution of $120\mu\text{m}$, below the size of a vessel (40 to $100\mu\text{m}$ for alder).

Homogeneity of the impregnation

However, the histograms still convey information, the first being on the homogeneity of the impreg-

nation. When two materials with distinct attenuations are mixed, in a volume that is smaller than the spatial resolution of the instrument, the attenuation lies in-between the attenuations of the pure materials alone. The distribution of this mixed attenuation, as reported on a histogram, gives information on the homogeneity of the mixture. On the histograms of the four different samples of figure 1, one can see that the longer is the impregnation, the more homogeneous is the distribution of the PEG in the wood.

Fig. 4 Attenuation through the sample, from tomographs made with a polychromatic and a monochromatic beam. – (Th. Demoulin).



Spatial distribution of the attenuation

The segmentation of the tomograph in artificial colours allows a comparison between the samples, as illustrated in figure 3, where the lighter the colour is, the higher is the attenuation. It has to be noted that this segmentation has no relevance for the bark, as visible in the bottom of the sample impregnated for eight hours, and ten weeks, since it possesses another attenuation than the wood. The scheme in figure 3 in the left represents the cut of the sample after freeze-drying and the direction, symbolized by the arrow, from where the sample is seen in the tomograph.

The sample impregnated during eight hours has not been turned over in the bath and shows a preferential ingress of PEG by the top side (which is the left side in the figure). One can also see that the PEG did not reach the core of this sample. On the other hand, the samples impregnated during one and ten weeks have a higher attenuation and suggest that the PEG has fully impregnated the wood.

The sides of these samples show a higher attenuation than their cores, which suggests that the amount of PEG is higher at this location. It cannot be due to a migration of the PEG after cutting, because these sides have been cut before the impregnation process. This result raises the question of a possible artefact due to beam-hardening effect, i. e. a change

in the effective energy spectrum due to the dependence of cross-section with neutron energy. It would lead to an underestimation of the attenuation in the bulk of the samples. To test this assumption and evaluate the importance of the phenomenon, a tomograph using a monochromatic beam has been carried out. The attenuation profile through the sample is reported in figure 4. It clearly shows that the attenuation profile is similar using a polychromatic or monochromatic beam, and thus that the tomographs do not suffer from beam-hardening. By consequence, this higher attenuation is caused by a higher concentration of PEG at the surface.

It is important to point out that the value of attenuation that we get from the tomographs cannot be directly converted to a concentration of PEG in the wood, even by using the segmentation previously mentioned. Indeed, it is not known whether the impregnated wood has conserved the same density, and thus the same attenuation, as the non-impregnated wood. The segmentation allows a direct comparison but not a quantification of the concentration of PEG.

Efficiency of the treatment

Eventually, neutron tomograph allows assessing the occurrence of cracks, with dimensions larger

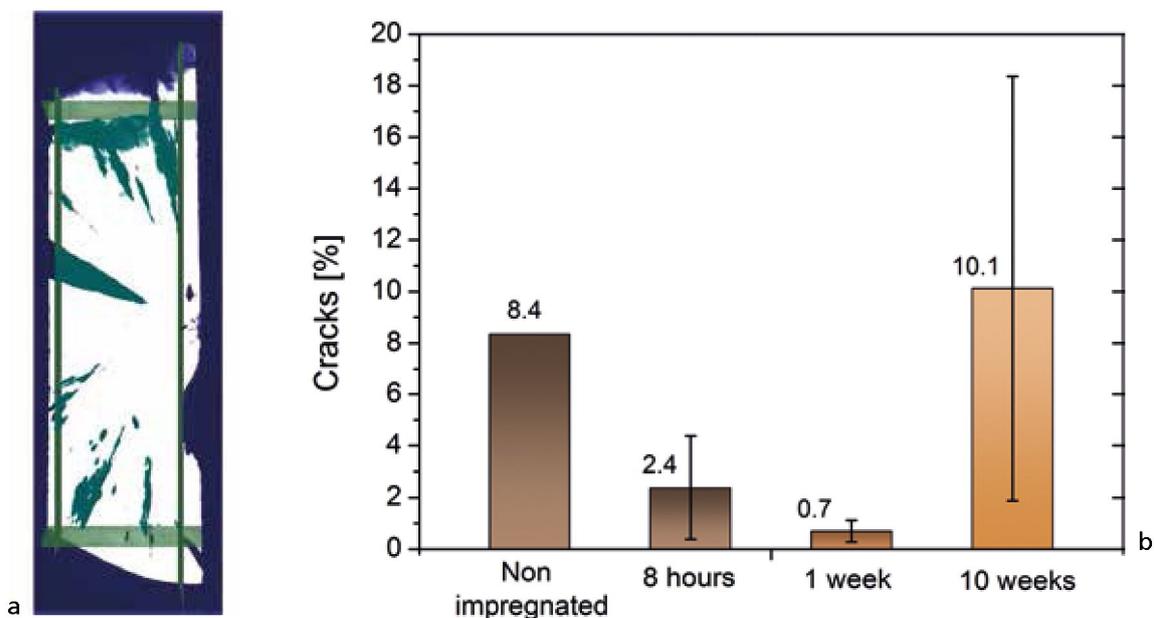


Fig. 5 **a** the cracks are visible in turquoise blue. The green box indicates the limit of the area for the quantification. – **b** amount of cracks expressed as a percentage of the volume of the sample. – (Th. Demoulin).

than the spatial resolution of the tomograph, after the conservation treatment. The separation between the air that constitutes the cracks, and the wood is made possible by the great difference in term of their respective grey-values. The cracks can then be visualized to see where they occur in the wood, and quantified to assess the overall efficiency of the treatment. In figure 5a are shown, in turquoise blue, the cracks during the process of quantification, and in figure 5b the amount of cracks for each series of samples. The sample, which has only been freeze-dried is in bad condition and 8.4% of its volume is made of air. The best-preserved samples, interestingly, are the ones impregnated during one week only, where the cracks represent 0.7% of the volume, while the worst ones are the samples impregnated during ten weeks. This would suggest that a long impregnation time and a homogeneous distribution of the PEG are not necessary to achieve a successful freeze-drying. However, the process of freeze-drying itself adds new variables and could be at the origin of these fractures, and therefore these results should be seriously followed-up.

Conclusion

This series of tomographs aims at interrogating the usefulness of neutron imaging for the study of conservation treatment of waterlogged wood, by impregnation with PEG and subsequent freeze-drying. First of all, the high cross-section of the PEG with cold neutrons limits the size of the samples to 2.5cm (with the energy spectrum of ANTARES in 2009), which restricts the investigations to samples that can be cut or to small wooden artefacts. Secondly, despite the fact that the spatial resolution does not permit (yet?) a direct discrimination between the wood and the PEG, neutron tomography proved to be useful for the visualization of the distribution of the PEG in the dry wood after the conservation treatment. However, a quantification of the concentration of PEG is not possible with the proposed segmentation. Finally, the same experiment allows evaluating the efficiency of the treatment by quantifying the amount of cracks and their location in the wood. The combination of these outputs on one measurement could bring useful hints on the conservation process.

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References

- Barbour, Archaeological Wood. Properties, Chemistry and Preservation. Advances in Chemistry Series 225 (Washington, D. C. 1989) 35-66.
- Bugani et al. 2008: S. Bugani / P. Cloetens / M. P. Colombini et al., Evaluation of conservation treatments for archaeological waterlogged wooden artefacts. In: 9th International Conference on NDT of Art, Jerusalem, Israel, 25-30 May 2008 (Jerusalem 2008). www.ndt.net/search/docs.php3?MainSource=65
- Grattan/Clarke 1987: D. W. Grattan / R. W. Clarke, Conservation of waterlogged wood. In: C. Pearson (ed.), Conservation of marine archaeological objects (London et al. 1987) 164-206.
- Grünauer 2005: F. Grünauer, Design, optimization, and implementation of the new neutron radiography facility at FRM-II [Diss. TU München 2005].
- Hoffmann/Jones 1989: P. Hoffmann / M. A. Jones, Structure and degradation process for waterlogged archaeological wood. In: R. M. Rowell / R. J. Barbour, Archaeological Wood. Properties, Chemistry and Preservation. Advances in Chemistry Series 225 (Washington, D. C. 1989) 35-66.
- Kaye/Cole-Hamilton/Morphet 2000: B. Kaye / D. J. Cole-Hamilton / K. Morphet, Supercritical drying: a new method for conserving waterlogged archaeological materials. Studies in Conservation 45, 2000, 233-252.
- Unger/Schniewind/Unger 2001: A. Unger / A. P. Schniewind / W. Unger, Conservation of wood artifacts: A Handbook (Berlin et al. 2001).

Summary / Zusammenfassung

Neutron Tomography of Archaeological Waterlogged Wood

When waterlogged wood is found, the main challenge for the curator is to dry the wood without deteriorating the object. It is often done by impregnating the wood with PEG (polyethylene glycol) followed by freeze-drying. The present work investigates the possibility of using neutron tomography to visualize the PEG in archaeological waterlogged wood after the conservation treatment. 18 samples of waterlogged alder, with dimensions of 2 cm × 1.5 cm × 7 cm, impregnated by PEG 1500 with different duration of impregnation have been used. If the PEG cannot be distinctly separated from the wood, its spatial distribution can still be visualized, but not quantified. Furthermore, owing to the large attenuation difference between the wood and the air, neutron tomography can be used effectively to study the location and the amount of cracks and thus the efficiency of the conservation treatment.

Neutronentomographie von archäologischem Nassholz

Wird wassergesättigtes Holz gefunden, ist es für den Kurator die größte Herausforderung, das Holz zu trocknen, ohne das Objekt zu schädigen. Dies geschieht oft, indem das Holz mit PEG (Polyethylenglycol) imprägniert und anschließend gefriergetrocknet wird. Dieser Beitrag untersucht die Möglichkeit, nach der Konservierung die Neutronentomographie zur Visualisierung des PEG in wassergesättigtem archäologischem Holz zu verwenden. Dazu wurden 18 Nassholz-Proben einer Größen von 2 cm × 1,5 cm × 7 cm verwendet und unterschiedlich lang mit PEG 1500 imprägniert. Selbst wenn das PEG nicht eindeutig vom Holz unterschieden werden kann, lässt sich seine räumliche Verteilung immer noch visualisieren, allerdings nicht quantitativ. Weiterhin kann aufgrund des großen Schwächungsunterschiedes zwischen Holz und Luft die Neutronentomographie effizient dazu genutzt werden, um Ort und Anzahl von Spalten zu untersuchen und damit die Wirksamkeit der Konservierung.

Keywords

waterlogged wood / neutron imaging / PEG / freeze-drying