Mediterranean Archaeology and Archaeometry, Vol. 13, No 3, pp. 137-143

NEUTRON IMAGING TECHNIQUES APPLIED TO STUDIES IN THE ARCHAEOLOGICAL AND CULTURAL HERITAGE FIELDS

M.A. Stanojev Pereira*, J.G. Marques, J.P. Santos, C.I. Burbidge, M.I. Dias, M.I. Prudêncio

Campus Tecnológico e Nuclear, Instituto Superior Técnico, Universidade Técnica de Lisboa, Estrada Nacional 10, km 139.7, 2695-066 Bobadela-LRS, Portugal

Received: 15/11/2012
Accepted: 02/08/2013
Corresponding author: mstanojev@ctn.ist.utl.pt

ABSTRACT

Neutron imaging techniques are non-destructive tools that can help to understand the processes involved in manufacturing and restoration of cultural heritage artefacts. The interaction of neutrons with matter is markedly different from the interaction of other types of radiation, enhancing features where light elements are involved. Imaging of relative levels of neutron absorption is based on photography of scintillations produced in a ZnS screen doped with LiF, where Li is used as a neutron to charged particle converter. In this work we present studies made in the Portuguese Research Reactor applied to the characterization of restorations with resin, absorption of natural oils, and water retention.

KEYWORDS: neutron tomography, scintillator, non-destructive, images, restoration, oils, authenticity

1. INTRODUCTION

Neutron imaging techniques (NIT), namely neutron radiography (2-D) and tomography (3-D), are important tools to the study of cultural heritage and the archaeological objects, since they allow the visualization of internal structure without compromising integrity (Deschler-Erb et al., 2004; Lehmann, 2005; von Der Hardt, Roettger, 1981).

As a complementary technique to X-ray and gamma-radiography, NIT fill an important gap in the study of the internal structure of objects, due to the fact that the neutron interaction with matter is different from the one of other types of radiation (Casali et al., 2009). The characteristics of this neutron-matter interaction make possible the visualization of substances containing light elements, such as oil, paraffin, water etc., even when these are surrounded by materials that would make them invisible under X-ray imaging (Rant et al., 2006; Kardjilov et al., 2006).

It is possible to verify, for example, restorations in ceramics and tiles, particularly where an organic adhesive was used, the retention of resins in ceramics following consolidation treatment, internal cracks in metal objects, structures related to the assembly of objects in materials such as bronze, iron, copper and wax statues, etc. (Prudêncio et al., 2011; Stanojev Pereira et al., 2012).

The aim of the present work is to illustrate the applicability of neutron radiography and tomography methods, for the evaluation of restoration and of the uptake/retention of liquids in contexts relevant to luminescence dating and conservation studies, which are of interest to archaeologists and cultural heritage scientists. Its objective is to show the utility of the method as a non-destructive tool for evaluation in authenticity testing, conservation, and archaeological reconstruction.

2. EXPERIMENTAL SETUP

The measurements reported in this work were performed at the neutron tomography facility of the Portuguese Research Nuclear Reactor (Stanojev Pereira et al., 2012). The Portuguese Research Reactor is a 1 MW pool-type reactor, mostly used for neutron activation analysis (Prudêncio et al., 2009; Dung et al., 2010). The tomography setup is installed at the horizontal access of the thermal column, a stack of graphite blocks, which provides a highly thermalized neutron spectrum (energy ca. 0.025 eV) at a flux of 2.2×10^5 n cm^{-2} s^{-1}. The main characteristics of the beam at the irradiation position are shown in Table 1 (Stanojev Pereira et al., 2012).

Table 1 - Characteristics of the neutron beam at the sample irradiation position.

Flux at irradiation position (*) (n cm^{-2} s^{-1})	2.2×10^5
Neutron spectrum	Thermal Maxwellian (0.025 eV)
n/gamma ratio (n cm^{-2} $mrem^{-1}$)	2.1×10^5
Resolution (μm)	391±10
Maximal beam diameter (cm)	5

(*)Au-foil method.

Fig. 1 shows a simplified layout of the neutron radiography setup: a neutron beam (a) impinges in a sample (b), and the transmitted neutron beam carries information about the sample when it hits on a scintillator screen (c). The screen is based on ZnS, a well known scintillator material that played an important role already in the discovery of the alpha-particle in the experiments of Lord Rutherford (Weber, 2002). In this case, we used a 0.42 mm thick NDg type based on ^{6}LiF/ZnS co-doped with Cu, Al and Au (Applied Scintillation Technologies, 2000; Stanojev Pereira et al., 2012), where ^{6}Li is used to convert neutrons into phosphor-exciting ions (Stedman, 1960). The image in the scintillator screen is reflected by a mirror (d) and captured by a Charge Coupled Device (CCD) camera (e). The camera is a Proline (Finger Lakes Instrumentation, USA), equipped with a Kodak KAF-1001E grade 1 CCD, with 1024 × 1024 pixels (24 × 24 μm size), (Eastman Kodak Company, 2001). A Peltier water cooling system is used to decrease the CCD temperature down to -65 ºC, to minimize its thermal noise. The mirror allows the placement of the camera outside the direct vision of the incident neutron beam, thus reducing the radiation damage on the CCD. A Nikon 50 mm/f1.4 lens was used in all images.

A 3-D image is achieved by combining multiple 2-D images with a known separation angle, 200 images in steps of 0.9° are used in this work for image reconstruction. This is an automatic process: at the end of the image capture at a given angle, the system sends an electronic signal to a rotary table (f), moving the sample for the next capture position and so on through the various angles (Prudêncio et al., 2011). Afterwards, the file containing all the 200 images is used by the software Octopus to perform image reconstruction and, the software VG Studio Max 2.0 provides a three dimensional view of the internal structure of the object (InCT, 2008; Volume Graphics, 2008).

The combination of neutron dispersion and alpha range (generated by scintillator screen), as well as sample/screen distance, optical dispersion of scintillation light, camera resolution and image processing results in a spatial resolution for tomographic imaging of (391 ± 10)µm.

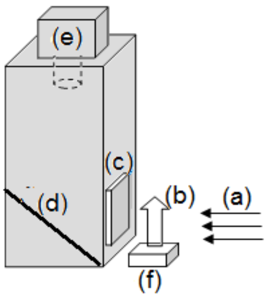

Figure 1. Schematic diagram of the setup for neutron tomography of the IST/CTN: (a) Neutron beam, (b) sample, (c) scintillator screen, (d) mirror, (e) camera, (f) rotary table.

3. CASE STUDIES

Two modern ceramic samples were chosen to demonstrate the applicability of this technique. The former is a small modern terracotta pot, acquired locally in Portugal (Fig. 2a) and the other one a replica Roman lamp acquired in Mérida (Spain) (Fig. 2b). These ceramic samples were specifically chosen because they are made of the material most commonly found in archaeological excavations. Their mineral composition is quite transparent to neutrons because it is composed primarily of silicates, which have low attenuation coefficient for thermal neutrons. However, the substances that are used in the process of restoration and conservation are rich in hydrogen in its structure, which makes ideal application of this technique (Munita et al., 2011; Rant et al., 2005).

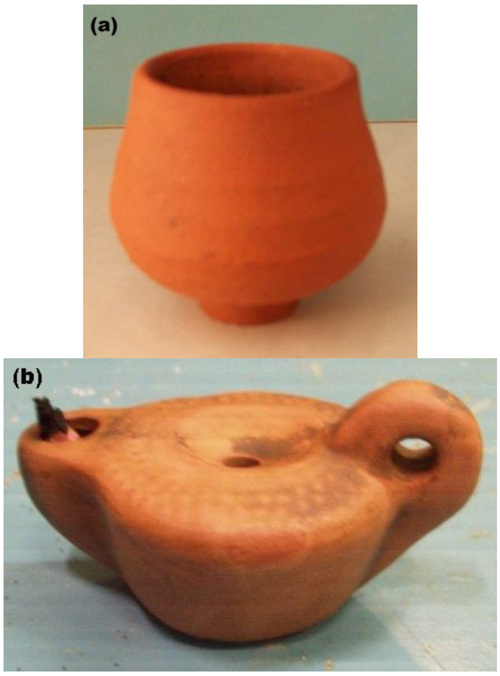

Figure 2. Pictures from the two ceramics studied: (a) pot, (b) lamp.

First, the 2-D and 3-D images of the untouched samples, without treatment of any nature, were acquired (Figs. 3a and 3b).

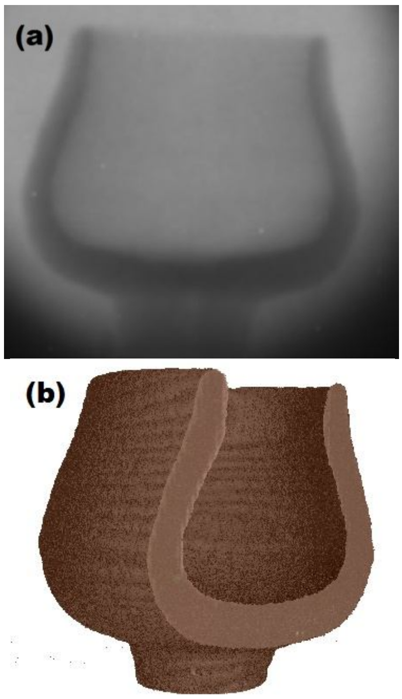

Figure 3 – Untouched pot image: (a) 2-D neutron radiography; (b) 3-D neutron tomography.

Second, the pot was broken and then mended by joining each of the broken faces using epoxy resin, and a new sequence of images was acquired (Figs. 4a, 4b, 4c and 4d). The aim of this study was to visualize marks and signals of the adhesive applied, perceptible to the neutron imaging technique, due to the high contrast between the materials composition of the pot (inorganic minerals; e.g. smectite $(Na,Ca)_{0.33}(Al,Mg)_2(Si_4O_{10})(OH)_2.nH_2O$) and the adhesive (organic; e.g. generic epoxy $C_{21}H_{25}ClO_5$).

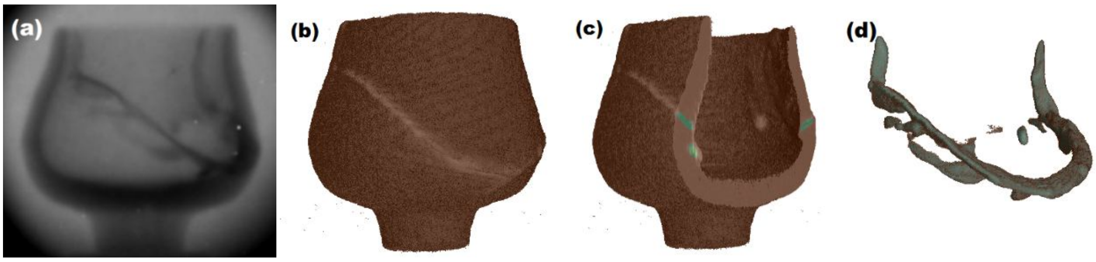

Figure 4 – Images of the broken pot restored: (a) Radiography; (b), (c) and (d) Tomographies, the latter is related to the adhesive structure.

Another phase of the work consisted on the study of the Roman replica lamp, where was inserted a wick made of cotton. As before, a series of untouched samples images were acquired (Fig. 5a and 5b).

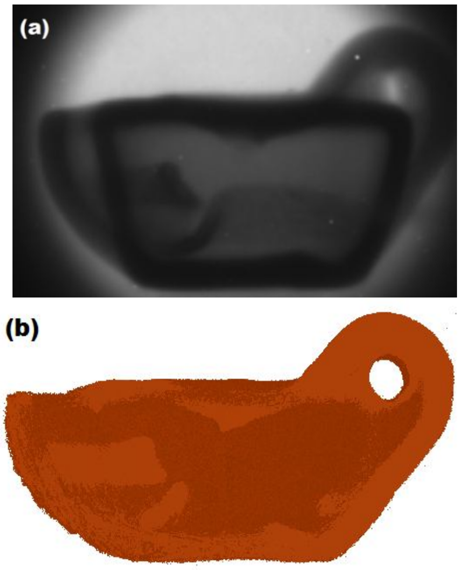

Figure 5 - Images of the untouched lamp. (a) 2-D radiography; (b) 3-D neutron tomography. In (a) darker areas, indicate a greater thickness of material, particularly around the edge of the piece. The wick is visible within, on the lower left hand side.

Next, olive oil was added to the lamp, and stored for 24 hours to permit impregnation of the wall of the object. Thereafter, the sample was left without oil for another 24 hours and the excess oil was dried with absorbent paper, and was irradiated again (Fig. 6a and 6b). This time, the purpose of the study was to observe the absorbed vegetable oil in the pottery and evaluate the capabilities of the technique in distinguish the three materials: pottery, oil and cotton.

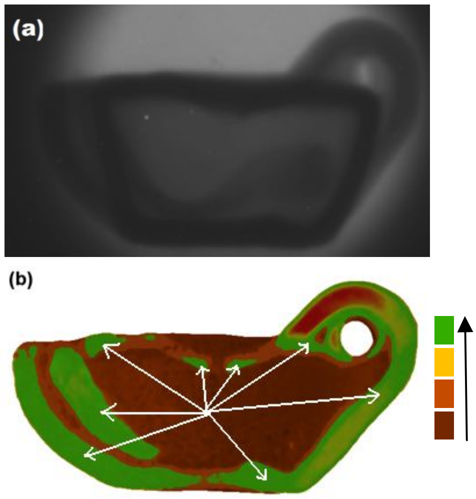

Figure 6 – Images of the lamp after insertion of olive oil. (a) 2-D Radiography and (b) 3-D neutron Tomography, where the arrows indicate the presence of the oil.

The sequence of images in Fig. 7 shows a test conducted to evaluate the potential for using NT in studies of water retention. One end of a Portuguese oak plank, 5 mm thickness, 35 mm height, was placed in a water reservoir (5 mm deep), and 2D neutron radiography images were obtained following different times to assess the evolution of water uptake. The digital images were processed with the free software ImageJ, using subtraction tools and a median filter to enhance the features of interest.

These show approximately saturating exponential increase in the height of the water column with time, as a result of capillary action

(Fig. 7). For longer uptake times, differences in the distribution and pathways of water uptake are evident: one area to the right of the plank appears not to be connected to the base by capillaries, while an area at mid-height on the left shows reduced water content with renewed higher levels of absorption above it.

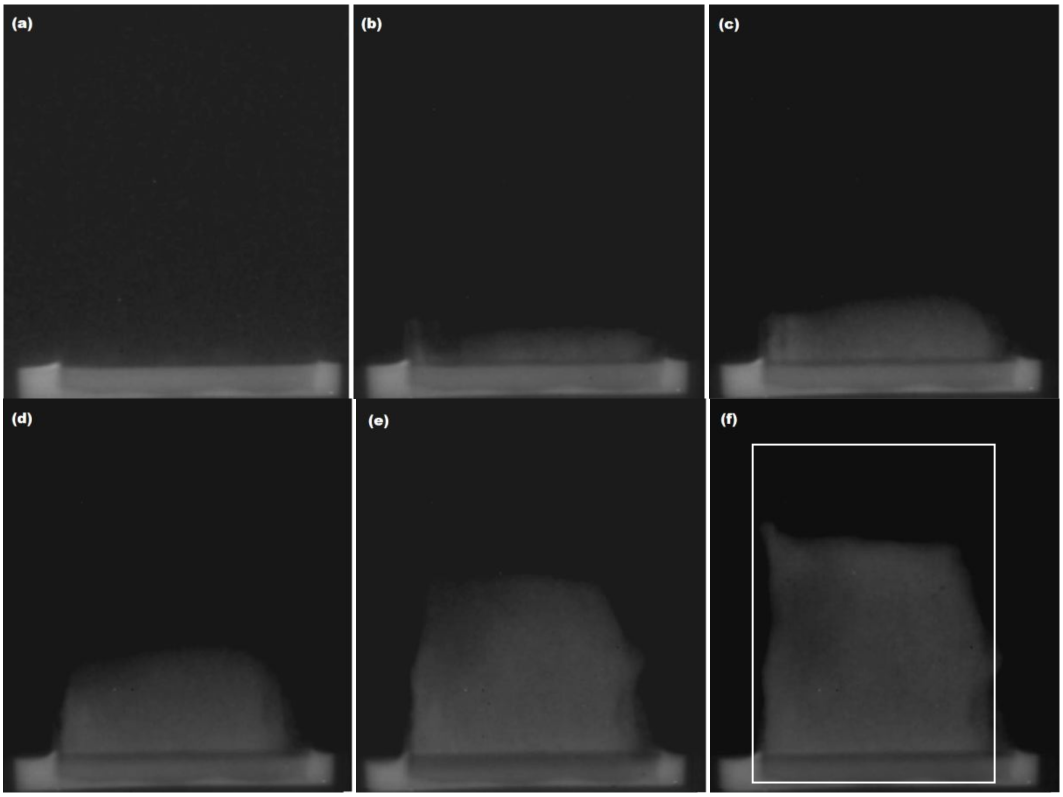

Figure 7 – Neutron radiography images of a plank wood in the water. (a) dry; (b) 20 minutes; (c) 40 minutes; (d) 90 minutes; (e) 180 minutes; (f) 330 minutes. The white line in (f) represents the limit of the wood.

4. DISCUSSION

Although the spatial resolution of this equipment is not ideal and the time required to acquire a neutron tomography is high (~ 1 h 40 min), the results obtained in this study were very promising.

Neutron interaction with the matter is nuclear, which means that for each atomic nucleus the neutron has a different behaviour. For example, hydrogen has a neutron attenuation coefficient of 23.2 cm^2 g^{-1}, and is the element to which the technique is most sensitive, while the value for lead is 0.032 cm^2 g^{-1} (von Der Hardt, Roettger, 1981). Thus, techniques that use neutrons to obtain radiographic images are complementary to those using electromagnetic radiation.

Because of that sensitivity to neutron absorption by hydrogen-rich substances, the technique was applied in studying a ceramic material, which has been restored using an organic adhesive, which composition enables detecting the presence of the substance, even in small quantities of material. Fig. 8 is the tomographic image section showing a thin film of adhesive with a 0.92(±0.02) mm thickness. The measurement tool is available in the software VG Studio.

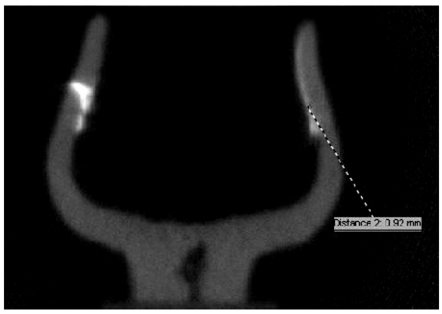

Figure 8. Neutron tomographic image of the pot ceramic slice.

In the same way, the technique was applied to the inspection of the impregnation of olive oil in a replica lamp. Through insertion of "pseudo-colour" in the tomographic image using the tool "components" of the VG Studio Software, the difference between the compared images of the lamp with and without oil was perfectly visible, though using grey levels its identification is impaired. The similarity in the grey level of the cotton wick, when comparing the images 5b and 6b, refers to sensitivity of the neutrons to the organic composition of the cotton. In this case the organic oil masks the organic cotton, but in the case of the wetted wood (Fig. 7a, b, d, e and f), the increase in absorption as the water content increases is clearly distinguished. Despite both wood and water being hydrogen-rich, in this case the mass of water absorbed by the wood is a significant proportion of the total.

The mapping of internal water in wood and ceramic artefacts is important for conservation because it relates strongly to various deterioration mechanisms (Prudêncio et al., 2011). The evaluation of water content is also important for environmental dose-rate reconstruction in luminescence dating (Aitken, 1985): the present work demonstrates how NIT can contribute to the evaluation of spatial variations in water content within artefacts. Small amounts of water or organics may be easily differentiated from a hydrogen-poor matrix based on their greater absorption cross-section, but even where absorption cross-sections are similar it is still possible to monitor differences, based on the mass of absorbing material. The present evaluations are qualitative: the development of calibration curves for evaluation of relative water content is underway, as previously conducted for resin in ceramics (Prudêncio et al., 2012). Fully quantitative evaluations will require calibration relative to independent measurements (changes in mass).

5. CONCLUSIONS

Techniques that use neutrons as a probe have proved to be a promising non-destructive tool for studying samples from archaeological sites and cultural heritage contexts, in particular for ceramics such as those studied in this work.

Restoration material is quite evident in Figs. 4a, 4b and 4c. In Fig. 4d, due to the software applied, the structure of the adhesive stands out from the ceramic structure so its shape and size can be quantified.

In Figs. 6a and 6b, particularly in the latter being a tomography image, the organic material can be clearly identified, visualizing the oil impregnated in the cotton wick.

Application of the technique in the mapping of water retention inside porous materials has been successfully conducted, and opens new perspectives for its application in materials of archaeological and cultural heritage interest, offering experts of these areas an important tool for inspection.

ACKNOWLEDGEMENTS

This work was supported by the FCT funded project PTDC/HIS-HEC/101756/2008 – RADIART.

REFERENCES

Aitken, M.J. (1985) *Thermoluminescence dating*. Academic Press.

Applied Scintillation Technologies. (2000) Phosphor/Scintillator Data Sheet 40, <http://www.appscintech.com/> (accessed July 31, 2013)

Casali, F., Morigi, M.P, Brancaccio, R., Montefusco, L., Jerjen, I., Flisch, A., Sennhauser, U., Liritzis, I., Tineanu, I. (2009) X-ray computed tomography for damage assessment of cultural heritage assets. *Prohitech-09 Conference (6th FP), Protection of Historical Buildings*, Rome 21-24 June (Mazzolani, ed), Taylor & Francis Group, London, ISBN 987-0-415-55803-7, 847-851

Deschler-Erb, E., Lehmann, E.H., Pernet, L., Vontobel, P., Hartmann, S. (2004) The complementary use of neutrons and X-rays for the non-destructive investigation of archaeological objects from Swiss collections. *Archaeometry*, vol. 46, 647-661.

Dung, H.M., Freitas, M.C., Santos, J.P., Marques, J.G. (2010) Re-characterization of irradiation facilities for k0-NAA at RPI after conversion to LEU fuel and re-arrangement of core configuration. *Nuclear Instruments and Methods in Physics Research A*, vol. 622, 438-442, doi:10.1016/j.nima.2010.02.057.

Eastman Kodak Company (2001) KAF - 1001E Performance Specification. Image Sensor Solutions, Rochester.

InCT, 2008. <http://www.inct.be/en/software/octopus> (accessed October 12, 2012)

Kardjilov, N., Fiori, F., Giunta, G., Hilger, A., Rustichelli, F., Strobl, M., Banhart, J., Triolo, R. J. (2006) Neutron tomography for archaeological investigations, *Journal of Neutron Research*, vol. 14 (1), 29–36.

Lehmann, E.H., Hartmanna, S., Wyerb, P. (2005) Neutron radiography as visualization and quantification method for conservation measures of wood firmness enhancement. *Nuclear Instruments and Methods in Physics Research A*, vol. 542 (1-3), 87-94.

Munita, C.S., Toyota, R.G., Neves, E.G., Demartini, C.C., Schaam, D.P., Oliveira, P.M.S. (2011) Chemical characterization of Marajoara pottery. *IAEA Radiation Technology Series (2),* http://www.iaea.org/Publications/index.html.

Prudêncio, M.I., Dias, M.I., Gouveia, M.A., Marques, R., Franco, D., Trindade, M.J. (2009) Geochemical signatures of Roman amphorae produced in the Sado River estuary, Lusitania (Western Portugal). *Journal of Archaeological Science* vol. 36(3), 873-883. doi: 10.1016/j.jas.2008.11.019.

Prudêncio, M.I., Stanojev Pereira, M.A., Marques, J.G., Dias, M.I., Esteves, L., Burbidge, C.I., Trindade, M.J., Albuquerque, M.B. (2012) Neutron tomography for the assessment of consolidant impregnation efficiency in Portuguese glazed tiles (16th and 18th centuries). *Journal of Archaeological Science*, vol. 39(4), 964–969. doi:10.1016/j.jas.2011.11.010.

Rant, J.J., Milic, Z., Istenic, J., Knific, T., Lengar, I., Rant, A. (2006) Neutron radiography examination of objects belonging to the cultural heritage. *Applied Radiation and Isotopes*, vol. 64(1), 7-12.

Rant, J.J., Milic, Z., Turk, P., Lengar, I. (2005) Neutron radiography as a NDT method in archaeology. *Proceedings of the 8th International Conference of the Slovenian Society for Non-Destructive Testing. Application of contemporary non-destructive testing in engineering*, Portorož, Slovenia 1–3 September, 181–8.

Stanojev Pereira, M.A., Marques, J.G., Pugliesi, R. (2012) A Simple Setup for Neutron Tomography at the Portuguese Nuclear Research Reactor. *Brazilian Journal of Physics*, vol. 42(5-6), 360-364. doi: 10.1007/s13538-012-0083-0.

Stedman, R. (1960) Scintillator for Thermal Neutrons using ^{6}LiF and ZnS(Ag). *Review of Scientific Instruments*, vol. 31, 1156; doi: 10.1063/1.1716833

Volume Graphics (2008). <http://www.volumegraphics.com/products/vgstudiomax/> (accessed October 16, 2012).

Von Der Hardt, P., Roettger, H. (1981) *Neutron Radiography Handbook*: Nuclear Science and Technology. D. Reidl, Dordrecht.

Webber, M.J. (2002) Inorganic scintillators: today and tomorrow. *Journal of Luminescence,* vol. 100, 35-45, doi: 10.1016/S0022-2313(02)00423-4.