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Neutron imaging of archaeological bronzes at the Oak Ridge national laboratory

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Abstract

This article presents the initial results of 2-D and 3-D neutron imaging of bronze artifacts using the CG-1D prototype beamline at the High Flux Isotope Reactor (HFIR) located at the Oak Ridge National Laboratory (ORNL). Neutron imaging is a non-destructive technique capable of producing unprecedented three-dimensional information on archaeomaterials, including qualitative, quantitative, and visual data on impurities, composition change, voids, and structure at macro-scale levels. The initial results presented in this publication highlight how information from neutron imaging can provide otherwise inaccessible details about the methods and materials that ancient craftspeople used in creating bronze objects.

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1. Introduction

Neutron imaging is a non-destructive, non-invasive technique capable of producing radiographic and tomographic data from a diversity of objects, spanning from engineering, geological, and biological materials to archaeological artifacts [1-5]. Unlike X-rays, which interact with an atom's electronic cloud, neutrons interact with the nucleus of an atom, where they can be scattered and/or absorbed. Their penetration through layers of metals is particularly well-suited for archaeological objects made of copper alloy.

In recent years a number of neutron-based techniques have been productively enlisted in the non-destructive two-dimensional and three-dimensional analysis of archaeological objects (e.g. radiography, Computed Tomography, diffraction) [6-9]. To date, the majority of neutron tomographic imaging of archaeological materials has occurred at major European spallation and pulsed neutron facilities, such as NEUTRA/SINQ at the Paul Scherrer Institute (Switzerland) and ISIS at the Rutherford Appleton Laboratory (U.K.) [1-2]. The neutron tomographic images collected by our research team at the Oak Ridge National Laboratory in August and May of 2011 mark the first time that archaeological objects have been examined with neutron computed tomography at a major pulsed neutron facility in the United States.

Neutron imaging, both two-dimensional (i.e. radiography) and three-dimensional (i.e. computed tomography CT), provides data on the structure, texture, phase contrasts, and composition of archaeological materials and objects at multiple scales and depths without requiring destructive or invasive sampling of the object. As demonstrated by the following Roman period examples of a small dog figurine and hanging oil lamp, such a holistic approach to rare bronze artifacts permits focus on a range of questions regarding manufacturing methods, craft skills, object integrity, and material behavior over time. These broad concerns are addressed with specific quantitative and qualitative data generated by the neutron imaging experiments. The data includes information on: material structure and heterogeneity; raw materials used in manufacturing; objects' functions and use-lives; and the relationship of structural and compositional variables to an objects' performance and preservation over time.

The goals of the 2011 experiments at the Oak Ridge National Laboratory were twofold and focused on testing the capabilities of the CG-1D neutron beamline for the effective imaging of archaeomaterials, and also on collecting initial structural data from a range of archaeological objects. A total of eleven artifacts were imaged in three dimensions, seven of which were bronze. The artifacts originate from contexts in the Mediterranean associated with Classical Greek and Roman settlement, or from early historical North American manufacturers and artisans in New England and the upper Midwest. All of the objects were loaned from the research collections of either the Joukowsky Institute for Archaeology and the Ancient World at Brown University or the Detroit Historical Museum in Michigan.

These initial measurements established a protocol for measuring archaeological objects at the ORNL CG-1D neutron imaging facility. Similar to the challenges faced with medical imaging, the research team worked to balance image noise with radiation exposure to minimize activation in the archaeological objects in order to ensure their safe and timely return to their institutional collections. The combined practical/simulation approach utilized for this research is described below.

1.1. Principle of conventional neutron imaging and application to Bronze artifacts

Neutron radiography is based on the attenuation of a neutron beam through the matter in which it passes. The decrease in transmission of the beam is caused by scattering and absorption within the object under study. Beam attenuation caused by a thick sample composed of a single isotope is given by Beer-

Lambert law

$$I(\lambda) = I_0(\lambda)e^{-\mu(\lambda)\Delta x} \quad (1)$$

where $I_0(\lambda)$ and $I(\lambda)$ are, respectively, the incident and transmitted beam intensities at wavelength λ , $\mu(\lambda)$ is the attenuation coefficient at wavelength λ and Δx is the thickness of the sample. The attenuation coefficient μ is given by

$$\mu(\lambda) = \sigma_t(\lambda) \frac{\rho N_A}{M} \quad (2)$$

where $\sigma_t(\lambda)$ is the material's total cross section for neutrons of wavelength λ , ρ is its density, N_A is Avogadro's number, and M is the molar mass. Attenuation coefficients are wavelength dependent, as described in Eq. 2. Straightforward extension of these basic formulas underpins the radiography of heterogeneous and irregularly shaped objects.

The process of creating a three-dimensional tomographic images first involves taking a set of radiographs of the object at multiple, evenly-spaced orientations. These radiographs collect projection data at the same time. The length of the imaging process depends on the desired accuracy and resolution, as well as the strength of the neutron flux. Image reconstruction (or computed tomography) from projections can be obtained using the Filtered-Back Projection (FBP) technique which produces sinograms. The intensities in the sinograms are proportional to the line integrals of the neutron attenuation coefficient between the source and the detector. The two-dimensional slice represents the attenuation coefficient distribution obtained by inverse Radon transform of these line integrals [11].

Metals pose multiple challenges in archaeometric research. Corrosion and poor preservation conditions often inhibit visual identification of objects and their manufacturing techniques. Heterogeneous constituents within archaeological metals, (related to both ore and alloy components), prevent accurate compositional and provenience analysis using techniques such as X-Ray Fluorescence (XRF) and Scanning Electron Microscope-Energy Dispersive Spectroscopy (SEM/EDS). X-ray imaging offers only limited penetration of metallic structures. Very few historical accounts describe the construction process of ancient metals. Although it is possible to obtain details about a metal object's manufacturing techniques, constituents, and properties from metallographic analysis, such examinations are invasive and destructive, since they require the removal of a small section of the object. Neutron imaging offers the possibility of performing metallographic studies non-destructively through neutron computed tomography.

The composition of bronze objects likely manufactured during the Roman period can vary considerably, but in general, ancient bronzes were composed primarily of Copper (Cu, 88%) and Tin (Sn, 12%), with trace amounts of Lead (Pb), Zinc (Zn), or other elements. The two samples discussed here, the dog figurine (JI-0000-04-0286, 286, Wagner Collection, Joukowsky Institute) and the hanging oil lamp (LC-025, Lewis Collection, Joukowsky Institute) are cast and molded examples of bronze objects. The dog sits 7.5 cm tall with a maximum width of 2.8 cm from hind leg to hind leg and possesses incised surface detail. The hanging oil lamp measures 17 cm in length and 8 cm wide. It hangs from braided bronze chains attached to decorative swan heads at the top of the lamp. Decorative lion heads protrude from either side.

As illustrated in Table 1, Cu, Sn and Pb scatter comparatively less than a strong scatterer such as H and do not absorb as much as ^{10}B , for example. Complementarily to X-rays, neutrons are capable of penetrating deep layers of Bronze objects and are thus an appropriate probe for archaeological bronze objects.

Table 1. Thermal neutron cross-sections of the main elements found in archaeological bronzes in comparison with Hydrogen (H) and Boron-10 (^{10}B).

Element	Scattering cross-section (barns)	Attenuation cross-section (barns)
Cu	8.03	4.5
Sn	4.89	0.63
Zn	4.13	1.11
Pb	11.12	0.17
H	82.03	0.33
^{10}B	3.10	3835

1.2. Experimental set-up

Neutron radiography and tomography were performed at the High Flux Isotope Reactor (HFIR) CG-1D beamline, which is located in the Cold Guide Hall of the facility. Fig. 1 illustrates the basic layout of a neutron imaging beamline. An aperture of diameter, D , is placed after the neutron source. The most straightforward layout involves the use of evacuated flight tubes positioned between the aperture and the sample to minimize neutron scattering from moisture in the ambient air. Size, location and shape of the aperture define the maximum spatial resolution achievable when flight tubes are utilized. The figure of merit of a neutron imaging beamline is L/D , where L is the distance from the aperture to the face of the scintillator (where the image is produced). Characterization of a beamline is defined by three properties of the instrument: (1) its flux at sample position, (2) the temporal stability thereof and (3) its divergence.

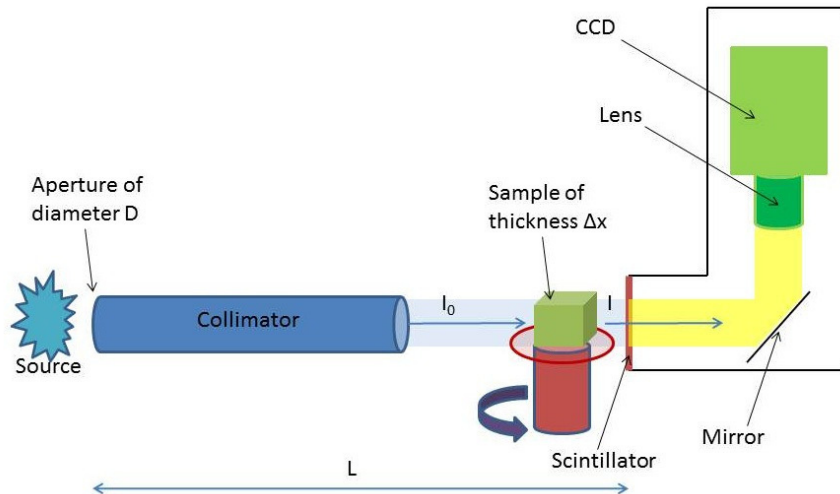


Fig. 1. Schematic layout of a neutron imaging beamline [12].

1.3. Protocol for estimating radioactivity in archaeological samples

The ORNL Neutron Sciences Directorate (NScD) has developed a tool called Sample Activation Calculation (SAC) that estimates neutron activation of a sample that has been exposed to a neutron beam. This tool is beamline-specific and complements experimental radiation measurements. The protocol employed for objects of cultural heritage at CG-1D has been as followed: (1) pre-exposure SACs using best sample composition estimation, (2) quick neutron beam exposure (30 sec or less), (3) radiation measurements using Geiger and neutron counters, (4) confirmation (or re-adjustment) of SACs based on experimental radiation measurements and (5) SACs for the duration of the estimated time required for computerized tomography (CT). This protocol ensures that archaeological objects, which belong to U. S. museums, can be returned within a year after neutron exposure. If an object is estimated by SACs to remain radioactive longer than a year, the archaeological team makes a decision to either limit exposure by performing radiography only or to balance CT optimization with radiation dose, similarly to diagnostic imaging in the medical community. The latter case may need adaptive statistical iterative reconstruction methods rather than the commonly used filtered back projection in order to reduce patient exposure time. Another approach to reduce neutron exposure may be the investigation and implementation, when possible, of these advanced medical CT techniques.

2. Results and Discussion

Two particularly interesting archaeological bronze objects have been measured using neutron radiography and CT at the HFIR CG-1D neutron imaging prototype facility. The achievable spatial resolution was approximately 75 microns. Fig. 2 displays the photograph and two-dimensional neutron-mosaic radiograph of a Late Roman hanging bronze lamp excavated from the site of Boscoreale, Italy. Although the lamp was too large to fit into the field of view and rotational platform used for three-

dimensional image collection on the CG-1D instrument, the research team compiled a photomosaic of two-dimensional radiographic images. The lamp's exterior is mildly corroded, but imaging revealed that it is otherwise structurally stable, with no superficial or subsurface cracks, voids or other weaknesses. Prior to imaging, it was known that lamp was constructed in a multi-part mold, but no other technical information was detectable about the application of decorative elements to the lamp's exterior, the joining of fasteners to the main body of the lamp, or how/if the lamp was used regularly in antiquity. The results clearly demonstrated the lamp's construction techniques, texture, and the presence of fuel residue and wear from burning in the lamp's interior. Future tests will examine the composition of the residue as well as the structural details of the joining mechanisms between the lamp's decorative elements and its main body.

Fig. 3 displays the photograph, neutron radiograph and neutron transmission contour map of an unprovenanced bronze dog figurine (probably Roman) from the Joukowsky Institute collections. The contour map indicates that transmission is lower inside the object, as expected since the object is thicker in the center. A CT scan of the dog figure was performed at CG-1D, as illustrated in Figs. 4. The tomography data identified defects such as voids inside the figurine (see Fig. 4b) and a change in attenuation values revealed faults in the manufacturing of the tail. The attenuation values of the tail are identical to those of the figurine's exterior layer. The internal structure of the dog figurine contains remains of the core material (clay and possibly organics), which indicate that the object was cast using a lost-wax technique. Future tests will examine the contrasts, composition, and variation between these internal features and the object's bronze exterior.

(a)



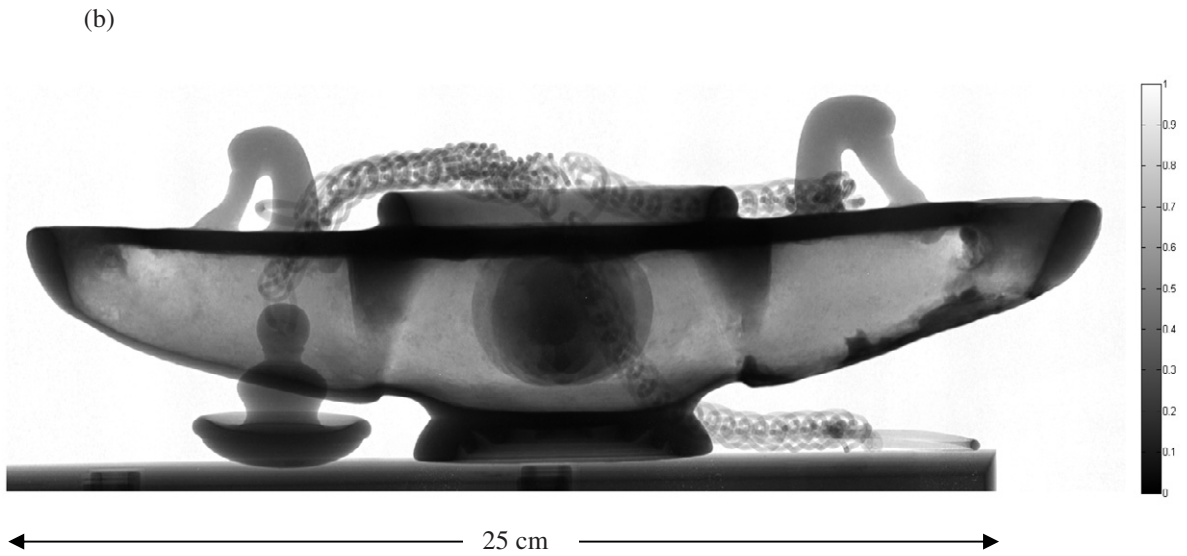


Fig. 2. Photograph (a) and 2-D neutron-mosaic radiograph (b) of Late Roman hanging bronze lamp showing inside residue. The transmission scale is displayed from 0 to 1 on the neutrograph, where 0 means 0% of neutron transmission and 1 means 100% neutron transmission.

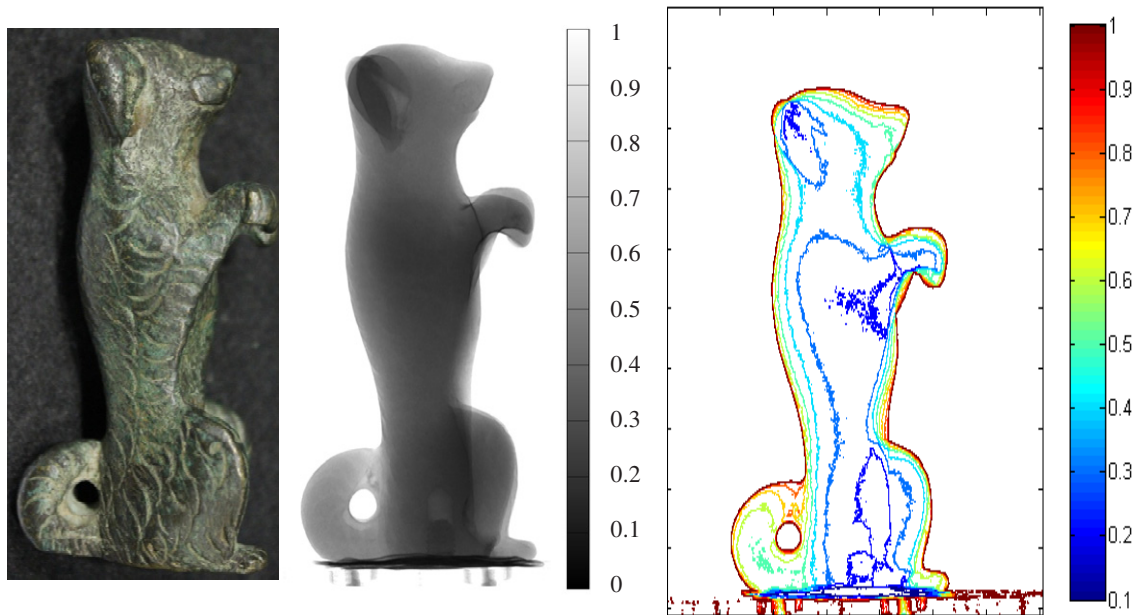


Fig. 3. (a) Photograph, (b) neutron radiograph and (c) contour map of an ancient dog figurine (7.5 cm x 3.5 cm).

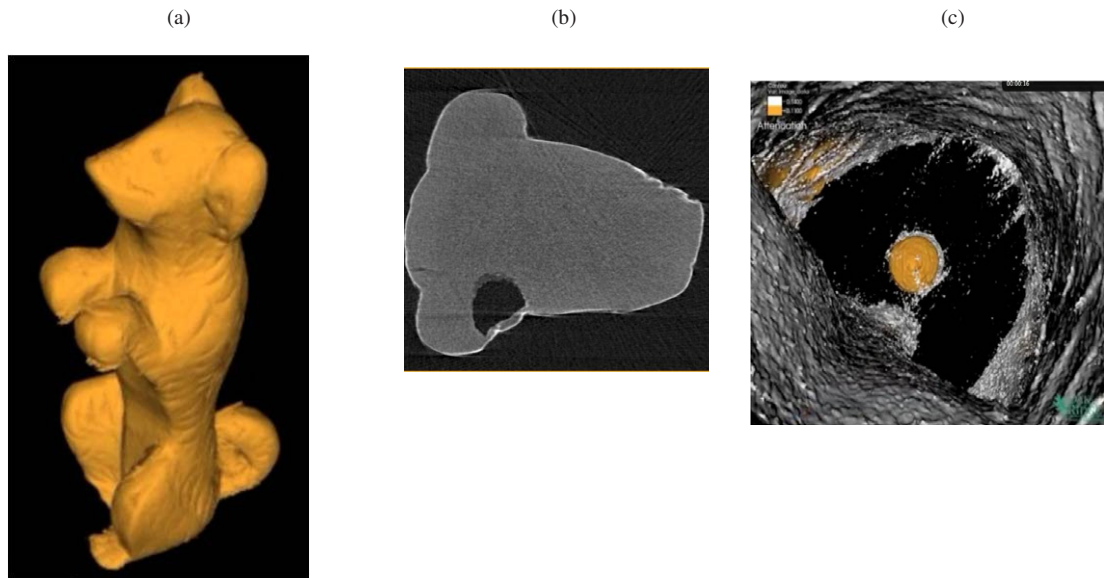


Fig. 4. (a) Visualization of 3-D CT scan of dog figurine, (b) neutron reconstructed slice of head of dog showing void in right ear, and (c) interior mid-section of dog figurine showing core material, internal material variation, texture and impurities.

3. Conclusion and Future Prospect

Two- and three-dimensional neutron imaging provides unique insights about archaeological objects without the necessity of invasively sampling a rare artifact. Nevertheless, careful planning during the experimental design stages is necessary to minimize neutron exposure, which leads to radioactivity in the sample. The results of our initial experiments, as illustrated by the hanging bronze lamp and bronze dog figurine, demonstrate the range of previously unobtainable information on material properties, structure, performance, and techniques of ancient craftspeople that can be obtained from non-destructive, non-invasive neutron imaging techniques.

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