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# Archaeometric investigations on manufacturing processes in ancient cultures with the neutron imaging station DINGO at ANSTO

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**Abstract**

This paper focuses on recent archaeometric investigations conducted with the neutron imaging station DINGO at ANSTO. The synergic application of non-invasive scientific analytical methods is becoming a common practice in archaeometry and conservation science. Neutron tomography is playing a significant role in expanding the technical limits and investigation capabilities of traditional analytical methods. We discuss advantages and limitations of the technique through the discussion of results obtained from the investigation of artefacts produced by different ancient cultures.


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*Keywords:* neutron tomography; non-invasive technique; archaeometallurgy

## 1. Introduction

The study of metal artefacts of archaeological, historical and cultural interest can shed light on the most advanced manufacturing processes developed by different cultures over time. Scientist need to treat them with care and must avoid damage, including the acceleration of any natural ageing process, so that we can pass on the artefacts to future

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generations.

The cultural heritage community is well aware of the benefits of non-invasive scientific methods. This approach has been progressively established as common practice in archaeometry and conservation science. Neutron imaging is playing a significant role in expanding the technical limits and investigation capabilities of standard analytical methods. This is due to its well-known features of high penetration power and different interaction with matter, compared, for example in the case of specific cultural heritage materials like metals and alloys, with the analogous X-ray imaging [Lehmann et al. (2011); N. Kardjilov et al. (2011); G. Bevan et al. (2013)].

While traditional analytical techniques might fail to preserve the integrity of the objects, neutron imaging methods can be successfully used to characterize the structure, morphology and composition of metal artworks three-dimensionally without the need for sampling or invasive procedures. These physical properties of an artefact are the imprint of its manufacturing process and of its life cycle. They can be convincingly reconstructed through a careful analysis of the material evidence.

In collaboration with museum institutions and university research groups, archaeometric investigations have been recently conducted by using the neutron imaging beamline DINGO at ANSTO [Garbe et al. (2015)]. A selection of case studies is presented in this paper focusing on the understanding of the processes used in the manufacture of a set of ancient metal artefacts through the characterization of their inner structure and morphology.

## 2. Methodological approach

The neutron tomography analyses were performed on DINGO, the ANSTO neutron imaging instrument located on a thermal beam tangentially facing the 20MW OPAL research reactor in Sydney [Garbe et al. (2015)].

Industrial processing, geoscience, palaeontology and civil engineering [Salvemini et al. (2016)] are the major applications in the utilization of the instrument. Additionally, research in the field of cultural heritage has also benefited from the use of neutron tomography. The flexibility of the DINGO layout allows the non-invasive analysis of samples of different sizes at the required spatial resolution, whose limit is imposed by the method and the current instrumental equipment.

The neutron tomography studies discussed in this paper were conducted in high resolution acquisition mode (length to diameter L/D ratios of 1000). In order to investigate large objects (case studies sections 3.1 and 3.2), a field of view of 200x200 $mm^2$ was used. This configuration features a pixel size of 100 μm and a 50 mm lens with a $^6$LiF/ZnS scintillation screen 100 μm thick was adopted. Projections were acquired every 0.25° over 360° with an exposure time of 15 s per projection. Since the length of samples exceed the actual size of the field of view available, consecutive tomographic scans, from 2 to 4, were acquired to investigate the entire artefacts. A superimposition of a few centimeters between scans allowed precise stitching of the individual tomographic reconstructions into a full 3D model of each sample during data processing. In some cases, it was not possible to investigate the whole volume due to the length of the sample and limited run of the samples stage along the vertical direction. An overall measuring time of about 12 hours was required per each scan.

A pixel size of 27 μm (case study 3.3) was obtained by setting a 55x55 $mm^2$ field of view with 100 mm lens, coupled with a $^6$LiF/ZnS scintillation screen 50 μm thick. In this configuration, projections were acquired with an equiangular step of 0.25°over 360° and an exposure time of 50 s each, resulting in a total scan time of about 20 hours.

The tomographic reconstructions were obtained by using Octopus package [Dierick et al. (2004)] while AVIZO [https://www.fei.com/software/avizo3d/] was used for data visualization, stitching and quantification.

## 3. Case studies

### *3.1. A forensic study on ancient Japanese swords*

Exceptional hardness, resilience and appearance make Japanese swords one of the most valued weapons of ancient and modern times [Yumoto (1958)]. These distinctive features derive from the quite unique manufacturing technique developed by sword-smiths. Historically shaped by the conjuncture of Japanese politics, economy and culture, the actual evolution of their manufacturing techniques is still not fully understood, especially aspects of metallurgical and metalworking practices. The method used by the early sword-smiths was never documented but rather kept secret and the necessary information was orally transmitted from the master to his most skilled pupils.

Neutron imaging and diffraction techniques have been demonstrated to be a valuable tool to qualitatively and quantitatively characterize, from macroscopic to atomic scale, material properties of such metal artifacts in a non-destructive way [Barzagli et al. (2014)].

This presented analysis is a forensic study of a set of *katana*, part of the East Asian Collection of the Powerhouse Museum - Museum Of Applied Arts and Sciences in Sydney. Origin, time period and authorship are known only for a group of samples by transliteration of the signature engraved on their hilt. While, according to stylistic study, only the time period can be assigned for a second group of blades, *mumei*, whose manufacturing tradition is still uncertain.

In the attempt to assign the manufacturing tradition of the un-known sample, neutron tomography and neutron residual strain measurements were performed. The data obtained from the well-known blades allowed us to build up a reference database of signatures, such as residual stress or diffraction peak width distributions, provided by neutron diffraction analysis, which are associated with particular manufacturing methods adopted by Japanese swordsmiths and therefore to be used as benchmark data that can be cross-matched with the results obtained from *mumei* samples.

Although a number of different blades were analyzed, in the current work we report only the tomographic analysis on blades which were classified as *katana* (Table 1), while the neutron diffraction data [Salvemini et al. (2016)] and results on *wakizashi* will be published separately.

Table 1: Swords from the East Asian Collection of Museum of Applied Arts and Science (MAAS).

| Inventory number | Period | Province | Author |
|---|---|---|---|
| H1360 | 1346 - 1370 | Bitchu | Sadatsugu |
| H5378 | 1661 - 1673 | Iwami | Tsuguhiro |
| H4839 | c. 1800 | Bizen | Yokohama Sukenaga |
| H6856 | c. 1600 | - | *mumei* |

The tomographic reconstruction highlighted different macroscopic structural features for the blades investigated (Fig. 1) in terms of distribution and amount of slag inclusions, porosity, defects and extension of the quenched area at the cutting edge. Based on the different attenuation coefficients, it is possible to virtually isolate the contributions of iron, inclusions and pores by applying a threshold-based segmentation method which consists in partitioning pixels depending on their intensity value [Russ et al. (2015)]. Then the structural components can be isolated and quantified (Fig. 2) for each sample (Table 2).

Sample H1360 shows the lowest percentage of metal and the highest relative amount of inclusions and porosity. This might suggest that the blade was extensively used and repeatedly polished. A significant portion of steel was removed during refurbishment which is not surprising considering this is the oldest blade among the samples investigated. Moreover, the diffuse porosity and presence of inclusions are indicative of low quality production.

The steel used for H5378 was poorly refined as well. Pores and inclusion are localised at the back of the blade and run along the profile of the welding lines. In fact, according to literature [Kapp et al. (1998); K. Nagayama (1997)], Japanese blades were made starting from a selection of steel lumps that were strongly pre-treated to obtain a homogenous and purified multilayered sheet. Distinctive carbon steels, characterized by different hardness, were shaped and welded in specific areas of the blade to optimize the mechanical feature of the final product.

Contrary to sample H5378, much more care was spent in the preparation of H4839 where the neutron tomographic reconstruction evidenced the presence of an area of detachment between cutting edge and body of the blade. This defect is probably a consequence of the special thermo-mechanical treatment the edge receives in the production process with typical water quenching of the edge and formation of martensite [Kapp et al. (1998)].

Finally blade H6856 is characterized by a low amount of porosity and by inclusions, which indicate medium quality production.

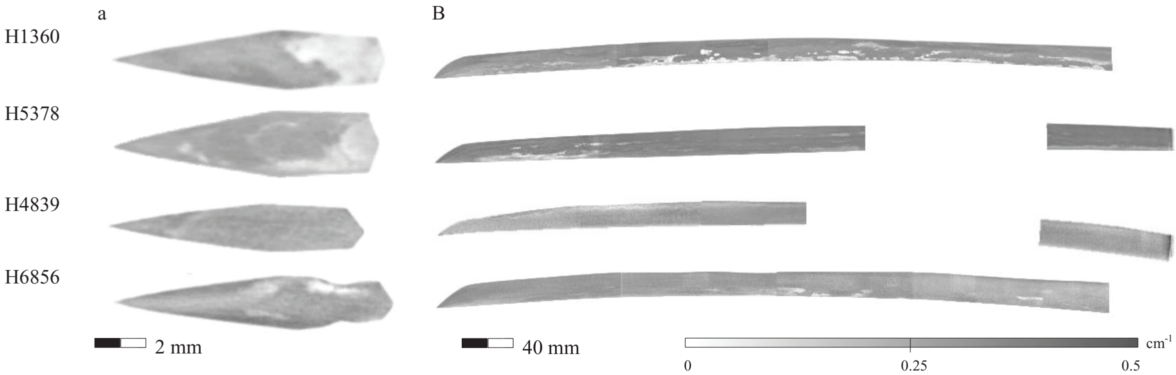


Fig. 1. Cross sections along the xy plane a) and the yz plane b) of the tomographic reconstruction of the four blades. At the bottom-right corner, the bar indicates the correspondence between grey tone and linear attenuation coefficient.

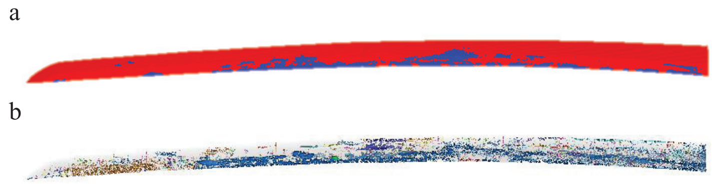


Fig. 2. a) A cut through the tomographic reconstruction of blade H1360; the red area is steel, while inclusions are colored in blue. b) 3D map of porosity rendered in false color.

Table 2: Quantification of percentage of pores, slag inclusion and steel evaluated over the investigated volume of the blades. The corresponding percentage error is also reported.

| ID | Porosity*(%)* | δ(%) | Inclusions*(%)* | δ (%) | Steel*(%)* | δ (%) |
|---|---|---|---|---|---|---|
| H1360 | 2.5 | 1.1 | 22.4 | 0 | 75.1 | 0 |
| H5378 | 8.1 | 0.1 | 7.1 | 0.2 | 84.8 | 0 |
| H4839 | 0.8 | 5.7 | - | - | 99.2 | 0 |
| H6856 | 2.1 | 1.3 | 3.5 | 1.3 | 94.4 | 0 |

The bulk macrostructure of *katana* characterized by neutron tomography can contribute to the information about the extent of refinement of the raw material and the care spent in the production of the blade; This data can be complemented by a microstructural characterization of the samples by exploiting complementary methods, such as neutron diffraction techniques, to attribute the specific blacksmith tradition of unknown blades.

*3.2. An authentication study of a bronze wine vessel from the China Shang Dynasty, 1600-1046 BCE*

The Chinese aristocracy of the Shang (~1600–1050 B.C.) and Zhou (~1050–221 B.C.) dynasties commissioned the casting of sumptuous vessels in bronze for making ritual offerings of food and wine to their esteemed ancestors. The technical sophistication required to produce these vessels attests their importance as symbols of power for the ruling class. During the Shang Dynasty, the Chinese developed a multi-stage process for casting bronze in sections of molded clay, known as lost-wax casting. First a model of the finished vessel was carefully carved in fine moist clay and then allowed to dry. The outer mold sections were produced by building up moist loess clay around the model; this outer layer was then cut away in sections, fired and reassembled for pouring of the molten metal. Depending on the shape of the vessel being produced, the mold assembly needed one or more cores, also of clay, in addition to the outer mold parts. Metal spacers, or chaplets, were carefully placed between inner cores and outer mold parts to maintain the proper distance to create the desired vessel thickness. Vessels with legs were cast upside down so that the legs served as inlets for pouring the alloy of copper, tin and lead [Avril and Bonadies (1991)].

A large number of these ritual artefacts, deposited as grave goods, were excavated from the tombs of the nobility and their burial contest is well-documented. However, a consistent percent of such artefacts was acquired by museums with very little record about their past, and historical accounts of deception and forgeries teach us that their authenticity might be questionable.

This is the case of a bronze wine vessel part of the East Asian Collection of the Museum of Applied Arts and Sciences in Sydney. In order to determine the manufacturing technique used in the production of the vessel and to assess its authenticity, the artefact was investigated by neutron tomography. The object is 200 mm height, 175 mm width and 90 mm depth (Fig.3) and, based on stylistic analysis, it is attributed to the Shang Dynasty accordingly to the acquisition information.

The neutron tomographic reconstruction of the studied vessel (Fig. 3) suggests the application of a traditional casting technique for the manufacturing of the vessel. Structural and morphological features of the artefact appear quite uniform in the bulk. Any sign of welding or soldering would have been detected at the interconnection of the structural components, but there was none. Traces of a repair attempt, already visible to naked eye, are only present where the vessel is more fragile, the protrusive rim. The analysis of the porosity (Fig. 3, b) showed a diffuse distribution of small tapped pores, indicating a good knowledge and high control of the casting process [Avril and Bonadies (1991)].

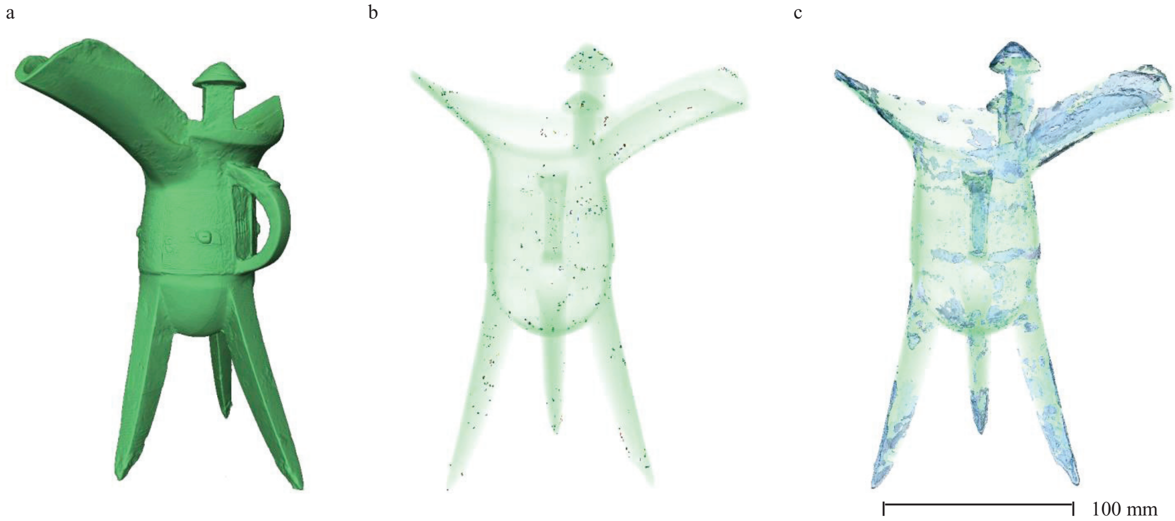


Fig. 3. a) 3D tomographic model of the sample; b) volume map of porosity; c) visualization of corrosion products (blue) diffusing into the bulk (transparent rendering).

Finally, important information on the state of conservation was obtained from the determination of the corrosion products that can be easily mapped due to the difference in neutron attenuation coefficients between metal structure and mineralization phases (Fig. 3, c). Corrosion is more evident inside the rim and at the base of the legs. Although areas of corrosion can be observed visually, the neutron tomography allows to judge about extent of corrosion products into depth.

Additionally, a neutron activation analysis was performed to detect if long–lived isotopes were naturally present in the artefact in order to prevent the risk of sample activation induced by long exposition to the neutron beam, sensitive aspects for cultural heritage artefacts that need to be returned to the museum exhibition within a reasonable period of time. Additional proof of authenticity of the vessel can be provided by complementary ion beam analyses that has been undertaken in order to characterize the elemental/isotope composition of the alloy. These results will be published separately.

### 3.3. The incuse coinage study

Coinage was first invented in the kingdom of Lydia in Asia Minor (modern Turkey) at the end of the 7th century BC. The use and production of coins then spread across the Aegean to Greece [Peacock (2006); R. Wallace (1989)]. In Southern Italy (Magna Graecia) the cities of Greek colonists independently developed a unique technique for the manufacture of coinage, known as the incuse technique. Incuse coins show the same image symbolizing the city of production on the front (obverse) and back (reverse) (Fig. 4, a). While the image on the obverse is shown in relief that on the back is sunk into the metal so that it appears as a negative or incuse version of the identical motif. The manufacturing processes need to create this coinage are poorly understood [Kraay (1976)].

In a collaborative project with the Australian Centre for Ancient Numismatics Studies (ACANS) at Macquarie University in Sydney (AU), silver coins from the ancient Greek colonies produced in South Italy during 6-5th century B.C. were investigated. In order to support comparative numismatic research on the subject of this unusual currency, specimens from different mints produced in different periods were chosen for study using neutron tomography in the hope that it might provide evidence for the nature of different manufacturing process (particularly incuse and non-incuse). The reconstructed volume of the samples permitted a quantitative investigation of the morphology and porosity of the metal. It also allowed the identification of inclusions and composite structures (Fig. 4, b). The detection and quantification of these features can provide information regarding the manufacturing technique and, in some case, can be a proof of authenticity. For example, neutron imaging was able to identify (unexpectedly) plated coins, due to the difference in attenuation coefficients between the copper-based material typically composing the core and the wrapping silver lamina. The components can be segmented, the respective volume estimated and the thickness of the lamina mapped in detail [Salvemini et al. (2016)].

a b c

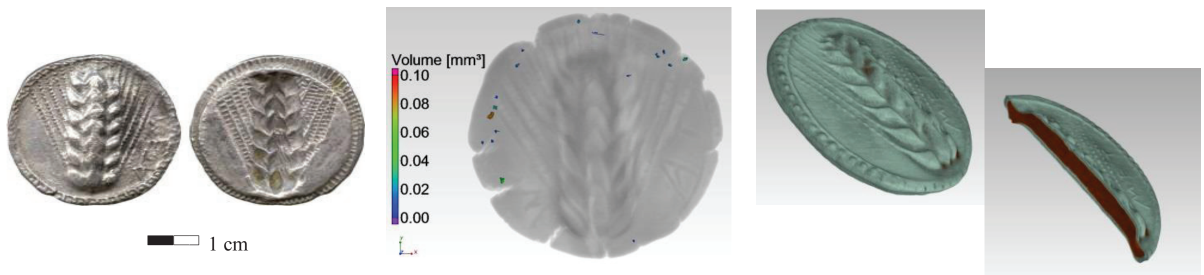


Fig. 4. a) A photograph of the incuse silver coin from Metapontum, South Italy (ACANS inv. 07GS526); b) Volume distribution of pores visualized inside the tomographic reconstruction of Metapontum coin (ACANS inv.07GS525); c) Tomographic reconstruction of a plated coin from Metapontum (ACANS inv. 07GS527) with green colored silver sheet and brown copper core visible from a cut through the sample.

Neutron tomography was combined with texture analysis performed on the neutron strain scanner KOWARI [ Kirstein et al. (2008)] at OPAL and with high resolution powder diffraction phase analysis on ECHIDNA [Liss et al. (2006)]. By investigating the orientation distribution of the polycrystalline structure of the samples, future study will provide a more in-depth understanding of different methods of minting.

## 4. Conclusions

In the light of these case studies, neutron imaging proves itself to be a powerful tool for the investigation of cultural heritage objects in a non-destructive and non-invasive way. This is especially important in examples of metal objects, similar to those reported here, which are usually not suitable for more traditional X-ray tomography. Qualitative information about the structure and morphology of the sample was provided through a visualization of the interior of the object from the tomographic reconstruction. In addition, the ability to isolate and quantify structural elements makes this method advantageous as a means to extract important information about the manufacturing processes used in the making of ancient artefacts and as a means to determine their state of conservation. When the authenticity of the artefact has been questioned, the tomographic data were used to provide answers.

Given the current stage of technical development in neutron imaging, however, the support of complementary techniques remains critical in order to obtain a more comprehensive characterization of the sample material. In light of the importance of a synergic combination of non-invasive analytical methods, the Cultural Heritage project has been started at ANSTO in order to interface a whole range of nuclear methods to extract maximum information from an object without the need for sampling or invasive procedures [http://www.ansto.gov.au/ResearchHub/Bragg/CurrentResearch/ScientificProjects/CulturalHeritage/index.htm].

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