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Archaeometry at the PGAA facility of MLZ – Prompt gamma-ray neutron activation analysis and neutron tomography

E.J. Kluge[a,*], C. Stieghorst[b,*], F.E. Wagner[d], R. Gebhard[c], Zs. Révay[b], J. Jolie[a]

[a] Institut für Kernphysik (IKP), University of Cologne, D-50937 Cologne, Germany
[b] Heinz Maier-Leibnitz Zentrum (MLZ), Technical University of Munich, D-85747 Garching, Germany
[c] Archäologische Staatssammlung, D-85598 Munich, Germany
[d] Fakultät für Physik, Technical University of Munich, D-85748 Garching, Germany

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ABSTRACT

Instrumental neutron techniques such as *Prompt Gamma-ray Activation Analysis* and *Neutron Tomography* and their combination, are effective methods to obtain chemical compositions with good detection limits and visualize internal structures within a sample. As non-destructive analysis methods, they are especially suitable for the investigation of cultural heritage objects and are therefore attractive for the field of *archaeometry*. This article reports on the investigation of two ring-like Celtic burial gifts from the Bavarian region using these methods. In contrast to our initial presumption, the two rings were not made in the same way. Our results clearly show completely different compositions and internal structures.

1. Introduction

Neutrons, as neutral particles, penetrate deep into matter. In contrast to many other analysis applications, neutron-based ones are especially suitable for the investigation of valuable objects, since no mechanical and chemical sample preparation is required. Methods like *Prompt Gamma Activation Analysis* (PGAA) and *Neutron Tomography* (NT), are straightforward techniques for chemical analysis with good detection limits (Molnár, 2004) and for the visualization of internal structures. Thus, they can contribute to the field of *archaeometry*.

Thanks to the latest developments in neutron sources and digital signal processing, the capabilities of γ-spectrometry and neutron imaging (Schillinger et al., 2006) have significantly improved during the last decades. At the *PGAA facility* of the *Heinz Maier-Leibnitz Zentrum* (MLZ) it is possible to perform both (Kudějová et al., 2007; Schulze et al., 2013).

We report on the investigation of two ring-like Celtic burial gifts from the Bavarian region. In contrast to the initial presumption, the two rings were not manufactured in the same way. Our results show completely different element compositions and internal structures.

2. PGAA instrument

The *PGAA facility* with its flexible instrumental arrangement[1] is located in the *Neutron Guide Hall* of the research reactor FRM II in Garching near Munich (MLZ et al., 2015). The facility and its constantly evolving instrumentation is built and maintained by a collaboration of the MLZ and the Institute for Nuclear Physics of the University of Cologne (IKP) (Kudějová et al., 2005, 2008).

To raise the neutron interaction probability with the sample material and to reduce hard γ-radiation from the reactor, the facility is connected to a cold neutron source with a curved neutron guide. Subsequently, the beam gets further adjusted using a system of two, one-meter long remote-controlled units: 1) an ensemble of boron carbide collimators covered with lead shielding with apertures of 20 mm × 20 mm and a resulting thermal equivalent flux of about $2 \cdot 10^9/\text{cm}^2\,\text{s}$, and 2) an elliptically tapered supermirror-coated borofloat-glass guide with a cross section of 10 mm × 16 mm, yielding a thermal equivalent flux of about $3 \cdot 10^{10}/\text{cm}^2\,\text{s}$ to be used for high-flux PGAA. In addition the flux can further be adjusted by beam attenuators.

For maximum flexibility, the following instrument environment, i.e. target chamber, four axis precision motion table and shielding, is mounted on a carriage. The shielding itself is made of several moveable sections with recesses for PGAA collimators and a tomograph. The

* Corresponding authors.
E-mail addresses: ekluge@ikp.uni-koeln.de (E.J. Kluge), christian.stieghorst@frm2.tum.de (C. Stieghorst), fwagner@tum.de (F.E. Wagner), rupert.gebhard@extern.lrz-muenchen.de (R. Gebhard), zsolt.revay@frm2.tum.de (Z. Révay), jolie@ikp.uni-koeln.de (J. Jolie).

[1] It includes the main PGAA, an in-beam *Neutron Activation Analysis* (NAA), a *Neutron Depth Profiling* (NDP), a *Prompt Gamma Activation Imaging* (PGAI) and a *Neutron Tomography* (NT) setup.

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target chamber can be evacuated and accommodates samples up to $10 \times 10 \times 10$ cm^3. A Compton-suppressed HPGe detector equipped with an ORTEC DSpec-50 spectrometer acquires the γ spectra (Révay et al., 2015).

3. Tomography instrument

For the newly designed *tomography* setup, a boron carbide pinhole aperture is placed in the focal point of the beam, creating an artificial point source from where a conical beam emerges (Söllradl et al., 2015). Since the cone-beam must fully irradiate the sample at the maximum distance of about 800 mm from the aperture, the sample dimensions are limited to about 70 mm in hight and width (Fig. 1). For the necessary projection images, the sample is positioned via an aluminium holder on the precision motion table as close to the tomograph as possible.

The optical system of the tomograph consists of a 5.5 Mega-pixel CMOS[2] camera with a 100 mm focal length macro-lens whose field of view onto the neutron-sensitive scintillation screen[3] is adjustable from about 30 mm to 100 mm. The radiography performance of the system is determined by an *L/D* ratio (Schillinger, 2001) of about 250 and the 84 μm spatial resolution of the scintillator.

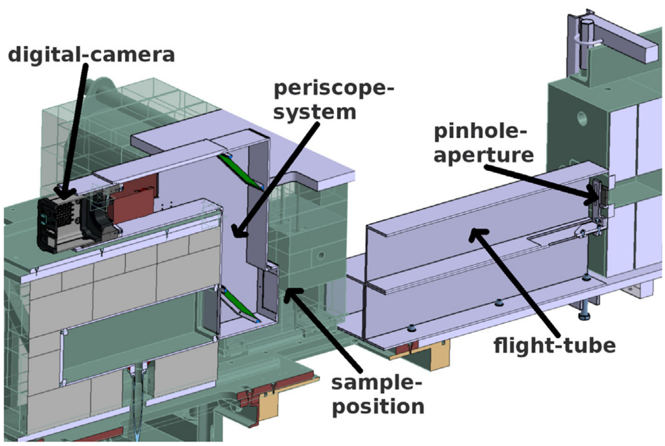


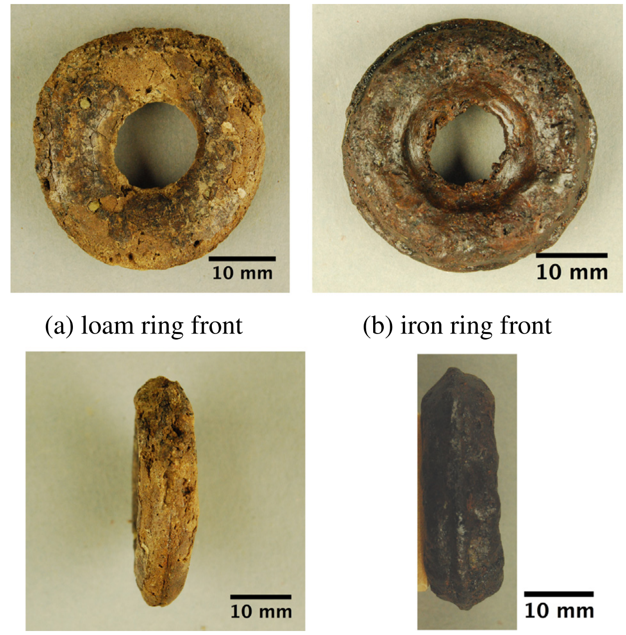
Fig. 1. CAD of the sliced cone-beam tomography setup.

4. Archaeological samples

Two similar ring-shaped objects (Fig. 2), found in different Celtic graves in Bavaria, were analysed via PGAA and NT. It is yet unknown, whether these amulet- or rattle rings, which are associated with the *La Tène culture* (Hoops, 2001; Riekhoff and Biel, 2001) and were also found during oppidum[4] excavations in south Austria and south-west Czechia, were solely used as burial gifts or had any practical application in life.

The first ring was found in 1889 at Huglfing, district of Weilheim-Schongau (Archaeological State Collection Munich inventory ID 1889.70). It visually consists mainly of loam with four symmetrically arranged tiny pins made of a different material (Fig. 3a, c) and weighs 10.595 g. It is hereinafter referred to as *loam ring*. The other, excavated in 1973 from the oppidum of Manching at Hundsrücken near Ingolstadt (Archaeological State Collection Munich inventory ID 1973.20), weighs 12.333 g and must, due to its weight-to-torus-volume ratio, only be coated with an iron shell that is corroded (Fig. 2b). It is hereinafter referred to as *iron ring*.

[2] Complementary metal-oxide-semiconductor.
[3] Al plate coated with 50 μm ZnS + ^{6}LiF(Ag).
[4] A fortified, town-like Celtic settlement from the La Tène period.



(a) loam ring front (b) iron ring front (c) loam ring side (d) iron ring side

Fig. 2. Pictures of the burial ring made of loam (a, c). Pictures of the fully iron coated burial ring (b, d).

5. Measurements and results

5.1. PGAA

The two objects were measured with *PGAA* using the collimator, different apertures and beam attenuations. The rings were fixed with FEP[5] strings in aluminium frames and positioned into the beam. The measurements were performed under vacuum. The loam ring was investigated with full beam flux and an aperture cross section of 10 mm × 2 mm at two different positions for about 60 min each. One position excluded any pins, the other included one (Fig. 3a). For the iron ring, the beam flux was attenuated to 7.5%, while its cross section was kept standard (20 mm × 20 mm). An arbitrary section, covering about 70% of the ring was measured for about 120 min.

Table 1 lists the most relevant element mass fractions found in the loam- and iron ring. In the loam ring, the elements H, B, Na, Al, Si, Cl, K, Ca, Ti, Mn, Fe, Cu, Cd, Sm and Gd were determined. The elemental compositions found in both sections – especially the high Al and Si concentrations together with Na, K, Ca and Mn – are in accordance with the presumption that the ring is mainly made of loam. However, the sections show a significant difference in the Mn and especially in the Cu content, suggesting the pins are corroded copper. Remainings of an outer crust, which is partially covering the loam, are also visible as darker areas in the X-ray image (Fig. 3a). This structure, in combination with the high iron content found, leads the assumption, that the loam ring originally may have been covered with an iron layer just like the other ring.

It was therefore reasonable to attribute the same structure to the iron ring. In the iron ring however, only the elements H, B, Si, Cl, Fe, Co and Cu were found. Since all main constituents of loam or clay except Si are missing, it is obvious that the iron coating is not forged or moulded around a loam or clay core. Moreover, the very low overall Si content of just 0.45(4) % implies the Si to be an impurity in or adherent to the iron.

[5] Fluorinated ethylene propylene.