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INSPECTION OF THE ROMAN TREASURE FIND BY NEUTRON-, GAMMA- AND X-RAY RADIOGRAPHY AND I-NAA

In autumn 2003 during the routine excavations on the construction site of the European highway Ljubljana-Zagreb (Croatia) near the village Drnovo the archaeologists unearthed an intact and excellently preserved Roman ceramic pot. The intact find, most likely containing a precious treasure, is an extremely rare event of world-wide importance. An opportunity to inspect intact Roman treasure that has been hidden for almost 1800 years is therefore a particularly exciting and very rare event for a specialist in non-destructive examinations (NDE).

Obtaining some preliminary knowledge of the contents of a find before their removal and conservation, such as the type and number of artefacts, their internal position and their material composition, is important for the archaeologist in order to elucidate the circumstances of the find, the historical time of the burial and to characterise each item. For a conservation specialist, the information obtained by non-destructive methods is important for properly emptying the pot, removing the artefacts and preparing for an appropriate conservation procedure.

Due to the rather extended dimensions of the pot (outer diameter ~20 cm) filled completely with soil it was a demanding task to radiographically examine the pot with X-rays, in particular with the 200 kV machine available at the National Museum of Slovenia. Therefore, the radiographic examination was carried out with thermal neutron radiography (NR) at the TRIGA Mark II research reactor in Ljubljana operated by the J. Stefan Institute and with the Ir-192 gamma rays of a private NDT company. Later the find was examined by X-ray computed tomography using medical CT device (120 kV) of the University Clinical Centre in Ljubljana. In the paper, following the description of the treasure find the various

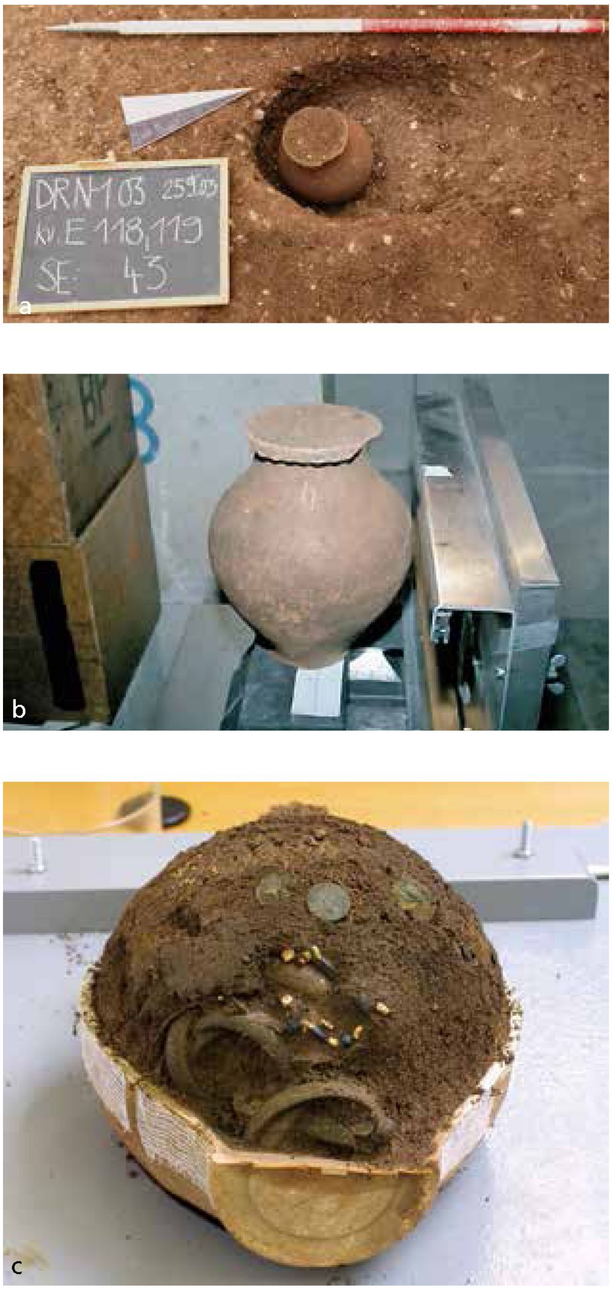


Fig. 1 **a** treasure pot during the excavation. – **b** unopened treasure pot set for inspection in the NR facility of the thermal column of Ljubljana TRIGA Mark II RR. – **c** the pot was cut by diamond saw to enable the removal of the contents.

neutron beam collimation (L/D ratio)	~ 65, range up to 80
useful beam diameter at detector	12 cm
beam shutter opening/closing time	~1 s
Φ_{th}-thermal neutron flux (Au foil)	~3.8 (range 3.4-4.6) 10^5 n/cm^2 *
R_{Cd} (Au)-Cd ratio	~9.6 (range 9.1-14.9) *
I_γ – gamma-ray intensity	~1.4 10^{-2} m Sv s^{-1}

Tab. 1 Characteristics of the Ljubljana TRIGA RR (250 kW) thermal column NR facility. – * Data depend on the reactor core configuration.

radiographic examinations are described and the results of the examination are reviewed. The Drnovo treasure find was extensively described in a monograph (Miškec/Pflaum 2007).

The Drnovo treasure find

The Drnovo treasure find was a ceramic pot completely filled with compact soil and earth (**fig. 1a**). The photography of the treasure find in the ceramic pot still unopened and positioned next to the neutron imaging camera is presented in **figure 1b**.
The total weight of the pot was 9.120 kg. The outer dimensions of the pot were:
height: 23.8 cm
diameter of the orifice: 13.3 cm
diameter of the bulge: 19.9 cm
diameter of the bottom: 9.0 cm
Almost at the same time another treasure find was unearthed nearby, although in this case the ceramic pot was destroyed by a farmers plough well back in the history and the contents, mostly silver coins, were found dispersed across the field. This led to the assumption that the intact pot that was found could possibly contain a treasure. The pot was dated to the second half of the 3rd century, later confirmed by the coins found in the pot and coins in the field.

Experimental work

Since the pot was a relatively thick object for investigation with the available 200 kV X-rays, the Ir-192 gamma-ray radiography and thermal neutron radiography (NR) were selected for examination of the pot's contents. The use of NR was also indicated in order to visualise possible remnants of any of organic materials, such as for instance textile or leather bags normally used to carry around money in ancient time.

Neutron radiography

The principles of NR, the experimental techniques and equipment have been described previously and can be found elsewhere (Domanus 1992).
In the past, the NR facility in the thermal column of the Ljubljana TRIGA Mark II reactor has already been used successfully for the NR examination of archaeological objects (Rant et al. 1994; 1996; 1997). Applications of NR to archaeology and to the examinations of objects of cultural heritage performed at J. Stefan Institute in Ljubljana have been reviewed recently (Rant et al. 2006).

NR facility of the Ljubljana TRIGA Mark II research reactor

Basic neutron radiographic data of the NR facility are presented in **table 1**. The neutron beam flux is rather low (~10^5 n/cm^2) in comparison with advanced NR facilities built around powerful neutron sources (e. g. SINQ at PSI, Switzerland or FRM II in Garching, Germany), there is a narrow useful beam diameter (~12 cm) and of medium neutron beam collimation. Nevertheless, the facility has been successfully used for NR inspection of archaeological objects during the past 20 years.

Digital neutron imaging system

Rather poor neutronic characteristics of the NR facility were compensated by substituting the rather slow film/Gd screen based direct neutron high resolution imaging technique by efficient direct digital neutron imaging using Gd doped imaging plate neutron detectors (IP-NDs) produced by FUJI Photo Film Co. The IP-NDs are read out by a FUJI BAS 1500 reader with 0.1 mm pixel size and 10 bit digitalisation. Use of FUJI IP-NDs reduced the exposure time from 1.5 h with the Gd screen/radiographic film method to only 20-100 s (factor 100), greatly increasing the speed of the NR examinations and reducing the activation of the object. In our case of investigating a rather thick pot filled with soil the exposure time was increased to 2-3 min.

NR examination procedure

The effective neutron beam diameter (~12 cm) does not cover the whole object and a complete imaging of the pot was obtained by a series of exposures. In two series of exposures, the neutron beam was centred once for the upper part and neck of the pot, and separately to image the lower part including the bottom of pot. In both series, the imaging of the central part of the pot (belly) was overlapping. In each of the series three irradiation positions were chosen by translating the pot for 4 cm from the left rim to the right rim and in each position the pot was examined in two positions rotated by 90 degrees. All together twelve neutron exposures were performed in a total duration of about 32 min. Almost complete information on the pot's contents was thereby obtained. More accurate information with grater image quality could be obtained by neutron CT using far more intensive and better collimated neutron beams of NR facilities e.g. at the Paul Scherrer Institute, Villigen (Switzerland) or the Hahn-Meitner Institute, Berlin. Unfortunately, the problems with the safe transportation and costly insurance of the very valuable treasure prevented such a thorough examination.

Activation of the object

Due to the low neutron beam fluence rate (~3.5×10^5 n cm^{-2} s^{-1}), the activation of the find was almost negligible. The total $\beta + \gamma$ doses rate on the surface of the pot and 5 min after the irradiation was 3-5 μSv/h and after 1 h approached the background value of 1.5 μSv/h. The precious find could hence be released back to the museum in the same day.

Instrumental neutron activation analysis

After the last neutron irradiation the pot was placed on the surface of HPGe gamma spectrometer, removing the Pb/Cu top shield. The counting time was 30 min. The analysis was purely qualitative, since the detector efficiency could not be estimated due to the complicated counting geometry and of the dimensions of the object.

Ir-192 gamma ray radiography

For Ir-192 gamma ray radiography a standard Ir-192 source of 7×10^{11} Bq (19 Ci) activity and 1.9 mm × 2.1 mm dimensions was used. The AGFA Structurix D7 radiographic films with standard lead cover were used. The FOD was 800 mm and film exposure times were 30 min. The image of the whole object was recorded. Again the object was rotated for 90 degrees.

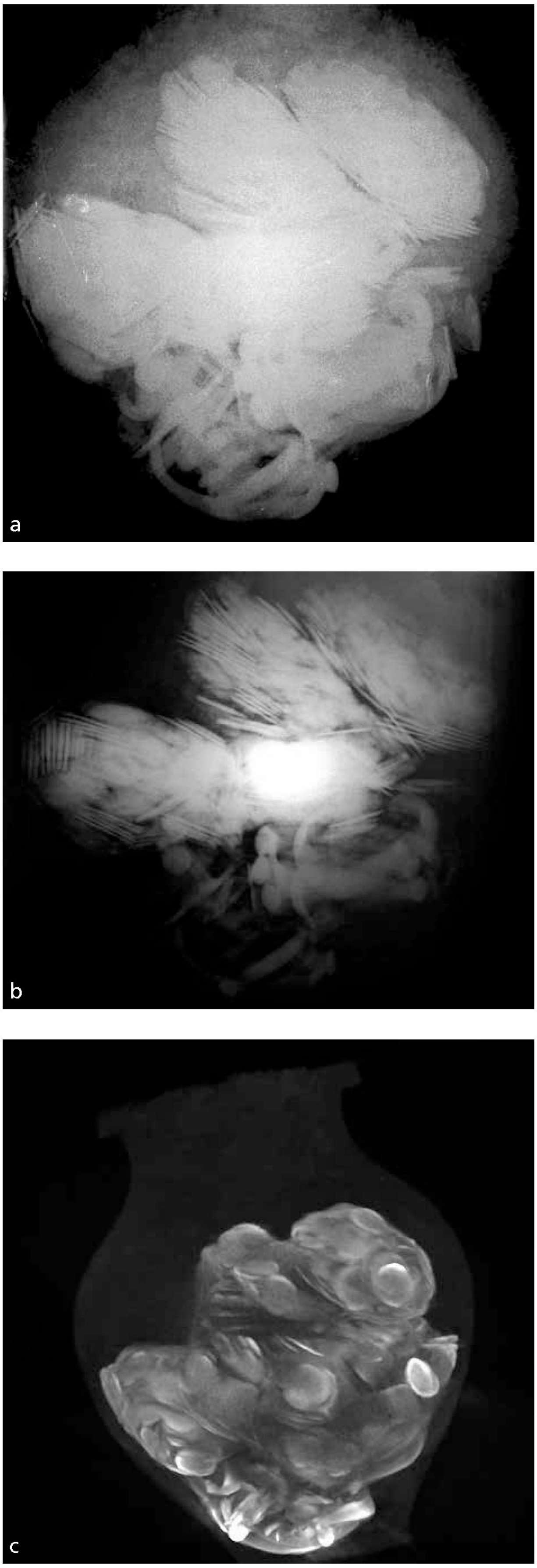
X-ray computed tomography

A medical X-ray CT (120 kV) unit of the University Clinical Centre of Ljubljana was used to obtain 3D image of the outer layers of the find.

Results of NDE

Neutron radiography

Neutron radiography (**fig. 2a**) revealed the position of four different groupings of coins, possibly contained in bags or sacks made of textile or leather. The coins could clearly be resolved only if they were located near the rim of the pot, where the exposure was sufficient to obtain a good contrast. The presence of the jewellery was revealed in the lower part of the pot and on the bottom. The presence of *fibulae* and bracelets was revealed.



Instrumental neutron activation analysis

The I-NAA revealed the presence of copper (from annihilation radiation and 1345 keV line of 12.8 h Cu^{64}) and gold (412 keV). In addition, activated common constituents of soil as Fe, Mn, Na, Ca and natural soil radioactivity (K^{40}, U and Th decay products) were detected. It was not possible to detect silver due to the short decay times of activated Ag isotopes, short irradiation time and after ~30 min of waiting time. Surprisingly, no golden artefacts were subsequently found in the pot and later analysis revealed the presence of gold as constituent element of an alloy in the silver jewellery. Since the presence of copper was confirmed, one could conclude that the hoard contains copper coins (*denarius*), possibly covered by silver.

Ir-192 gamma-ray radiography

The gamma-ray radiography (**fig. 2b**) clearly revealed that the money bags (purses) are filled with coins and that the coins are mostly of the *denarius* type. The presence of *fibulae* and bracelets was also clearly confirmed. However, again the object was too thick in the middle of the pot to allow visualizing and resolving of the contents in the middle section.

X-ray CT

X-ray CT in **figure 2c** revealed single coins in outer layers of the treasure and, in addition, confirmed the results of the NR and gamma-ray radiography.

Fig. 2 **a** neutron radiography. – **b** Ir-192 gamma radiography. – **c** X-ray CT of the central and lower part of the treasure pot revealed the coins being are hoarded in four distinct purses. Silver jewellery – three *fibulae* and eleven bracelets are located on the bottom of the pot.

	Fe	Ni	Cu	Zn	Ag	Sn	Au	Pb	Bi
fibula 1, bow	0.11		17.8	3.5	75.7	0.33	0.51	1.95	0.04
fibula 1, spot	0.51	0.06	35.6	6.0	35.8	18.9	0.52	2.79	
fibula 2, bow	0.02		15.3	3.4	79.0		0.44	1.86	0.06
fibula 1, spot	0.43	0.05	27.9	3.8	26.4	36.0	0.45	4.91	

Tab. 2 Results of the PIXE analysis (in %).

Determination of the composition of restored jewellery by PIXE

Using the proton induced X-ray emission (PIXE) four measurements of the element composition were made of a pair of silver *fibulae*, two on each. One measuring point was chosen on the bow of the *fibula*, and the other on a grey spot that visibly marred the smooth silver surface. The measurements were performed at J. Stefan Institute in Ljubljana (2 MeV proton beam, emitted into air through eight micrometres thick Al foil). The induced X-ray yields were detected with a silicon semiconductor detector equipped with an Al absorber of 0.1 mm thickness. The results are shown below (**tab. 2**). The accuracy of the major components is ~±5 %, and of the minor and trace elements ~±10-20 %.

At both spots on the bow of the *fibulae* silver with a purity of 76-79 % was measured. As a less precious metal, it contained somewhat less than 2 % lead, 15-18 % copper and 3.5 % zinc. The presence of zinc indicates that the silver was rarefied with brass.

Both grey spots could be noted to contain a significantly greater proportion of copper (28-36 %), an increased lead content (to 5 %), and a considerable proportion of tin (19-36 %) characteristic of the white bronze used in the Roman period to produce *fibulae* and other objects. In conclusion, the *fibulae* were not made from solid silver, but from a bronze with a high proportion of tin. The final product was coated with a thick layer of silver that was thriftily diluted with bras (Miškec / Pflaum 2007).

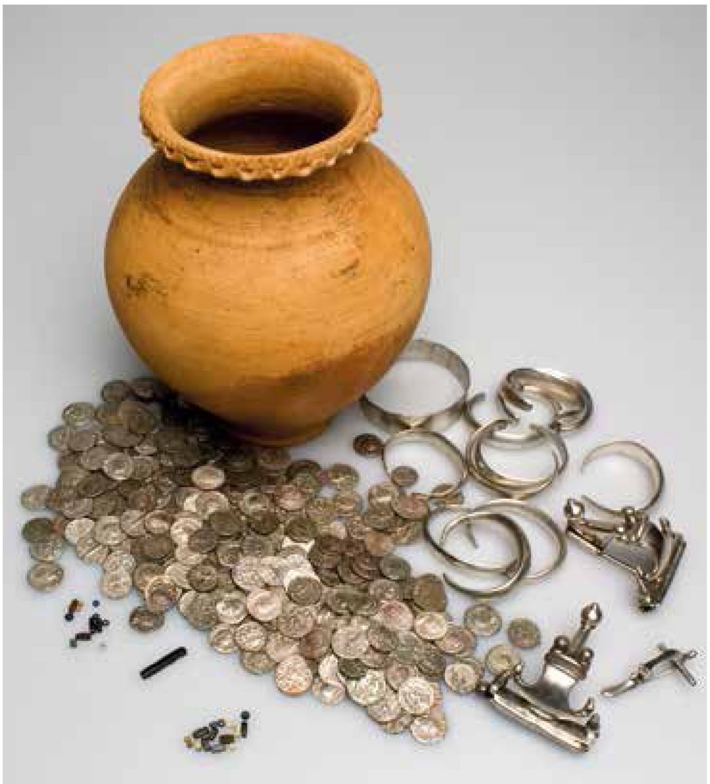


Fig. 3 The pot and its inventory after the restoration: coins, silver jewellery and beads of glass, stone and bone.

The inventory of the hoard

After the NDE provided rough information about the inventory of the pot and the relative position of the contents, the pot was cut by a diamond saw in two parts (to be later fixed together by a glue) and the contents carefully removed (**fig. 1c**). In addition to the coins that were wrapped in fabric, the pot also contained silver jewellery and numerous glass, stone and bone beads, which represented the remains of necklaces. The inventory of the find after the restoration is presented in **figure 3**.

Jewellery

The silver jewellery consisted of eleven bracelets and three *fibulae*. Ten of the bracelets were made of the solid silver in the form of an open ellipsoid hoop with tapering and conical ends. One bracelet was hoop-shaped silver bracelet. The weights of the bracelets differ: the lightest weighs 33.47 g, the heaviest 79.36 g. The open-ended bracelets can generally be divided into two groups, the first with a multi-angled section and the second with a semi-circular section. Graffiti (incised text) was found on three bracelets, and on one a decoration of hammered circles placed to form a cross.

Coins

The pottery vessel contained 973 Roman silver coins, consisting of 59 *denarii* while the others were *antoninianii*. The *denarius* was a silver coin that first appeared in the period of the Roman Republic and subsequently played an important role in the Roman Imperial monetary system up to the 3rd century when it was replaced by the *antoninianius*. The earliest coin in the find is a *denarius* of the Roman emperor *Septimius Severus* (ruled 193-211), while the latest were coins of *Gallienus, Postumus, Quietus and Macrianus II* from 260-261.

Fabric

The fabric used to wrap the coins partially adhered to the coins because of corrosion. In places it was torn or almost completely decayed. With consolidation and the careful removal of the coins, we succeeded in preserving a large fragment of fabric (13.9 cm × 11.7 cm) and several smaller fragments. Microscopic examination showed that the fabric was made of flax S-spun fibres woven in a simple linen pattern in which the woof thread interwove the warp thread. The count (the density of threads) in the preserved section was 18-thread/11-thread per cm^2.

Conclusions

The NR, Ir-192 gamma-ray radiography and X-ray CT usefully complemented each other in the visualising and characterising of the contents of the Drnovo Roman treasure find. The four groups of coins hoarded into the money bags made possibly of textile or leather were more clearly seen by NR, while individual coins were more clearly detected by gamma-ray radiography and X-ray CT. The positions of the bags and their contents were determined. Both methods were able to visualize several pieces of jewellery, consisting of three *fibulae* and eleven bracelets. The object was too thick to allow the examination of the pot in the middle section.

The I-NAA provided rough information about the material constitution of the pot's contents, confirming the presence of the copper, silver and gold. From this information and from the size of the coins it was concluded that the coins could be copper *denarii*. The presence of golden artefacts was not established, leading to the conclusion that the gold might be present in an alloy together with silver in the silver jewellery, as was subsequently confirmed by PIXE analysis.

NR and I-NAA proved to be useful NDE methods even at the small research reactor with modest neutron characteristics.

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Summary / Zusammenfassung

Inspection of the Roman Treasure Find by Neutron-, Gamma- and X-Ray Radiography and I-NAA

Towards the end of 2003 during the archaeological excavations near the village Drnovo on the construction site of the European highway Ljubljana-Zagreb (Croatia) an intact Roman ceramic pot assumed to contain a treasure find was unearthed. The ceramic pot was dated to the second half of the 3rd century. In order to obtain some preliminary information for the archaeologists and to properly conduct the opening and salvaging of the suspected precious contents of the find, non-destructive radiographic examinations using both conventional radiographic techniques (Ir-192 gamma ray radiography and X-ray computed tomography) as well as thermal neutron radiography (NR) and non-invasive Instrumental Neutron Activation Analysis (I-NAA) were performed. The pot was completely filled with earth, had a total weight of over 9 kg and was quite large (outer diameter 20 cm, height 24 cm) and hence presented a demanding task for radiographic examinations. The radiographic techniques provided clear evidence of the presence of a hoard of coins and jewellery and confirmed the assumptions about the treasure contents. The I-NAA gave a clue about the elemental composition of the hoard. The NR complemented the conventional radiography since it revealed that the coins were hoarded in the four separate purses made of organic materials, probably leather. The neutron NDE were performed using small 250 kW Ljubljana TRIGA research reactor demonstrating its capability for applications in archaeology.

Untersuchung eines römischen Schatzfundes mit Neutronen-, Gamma- und Röntgen-Radiographie sowie I-NAA

Gegen Ende 2003 kam während archäologischer Ausgrabungen in der Nähe des Dorfs Drnovo auf der Baustelle der Europastraße Ljubljana-Zagreb (Kroatien) ein unversehrter römischer Keramiktopf zutage, von dem man annahm, dass er einen Schatz enthielt. Der Keramiktopf wurde in die zweite Hälfte des 3. Jahrhunderts datiert. Um einige vorläufige Informationen für die Archäologen zu erhalten und um das Öffnen und Bergen des vermuteten wertvollen Inhalts korrekt durchzuführen, wurden zerstörungsfreie radiographische Untersuchungen sowohl mit konventionellen radiographischen Techniken (Ir-192 Gamma-Radiographie und Röntgen-Computertomographie) wie auch Neutronenradiographie (NR) mit thermischen Neutronen und nicht-invasive Instrumentelle Neutronenaktivierungsanalyse (I-NAA) durchgeführt. Der Topf war vollständig mit Erde gefüllt, hatte ein Gesamtgewicht von über 9 kg, war ziemlich groß (Außendurchmesser 20 cm, Höhe 24 cm) und stellte daher eine anspruchsvolle Aufgabe für radiographische Untersuchungen dar. Die radiographischen Techniken lieferten einen klaren Nachweis für die Existenz eines Horts von Münzen und Schmuck und bestätigten damit die Annahmen über den enthaltenen Schatz. Die I-NAA ergab einen Hinweis auf die elementare Zusammensetzung des Horts. Die Neutronenradiographie ergänzte die konventionelle (Röntgen-)Radiographie durch den Nachweis, dass die Münzen in vier getrennten Beuteln aus organischem Material – wahrscheinlich aus Leder – gehortet waren. Die zerstörungsfreien Neutronenuntersuchungen wurden an dem kleinen 250 kW TRIGA Forschungsreaktor in Ljubljana durchgeführt und zeigten damit dessen Eignung für Anwendungen in der Archäologie.

Keywords

archaeology / neutron radiography / Ir-192 radiography / X-ray CT / instrumental neutron activation analysis / Ljubljana TRIGA research reactor