Holistic documentation of Cypriot ceramic zoomorphic rhyta dating to the Hellenistic Period

Efstathiou[1], K., Georgiou, R.[1], Nikidiotis, T.[1], Ioannides, M. [1], Ganetsos, T.[2], Romantzi, K.[2], Papoytsidakis[2], M., Ioannou, L.[3], Kyriakou, M. [3], Hadjichristodoulou, C.[4] & Siegkas, P.[5]

[1] ERA Chair on Digital Cultural Heritage, Department of Electrical Engineering, Computer Engineering and Informatics, Cyprus University of Technology, Arch. Kyprianou 31, CY 3036 Limassol, Cyprus
[2] West Attica University, P. Ralli & Thivon 250, Athens, 12244, Greece
[3] Ygia Polyclinic Private Hospital, Nafpliou 21, 3025, Limassol, Cyprus
[4] Bank of Cyprus Cultural Foundation, Phaneromenis 86-90, 1011, Nicosia, Cyprus
[5] Department of Mechanical Engineering and Materials Science and Engineering, Cyprus University of Technology, Limassol, Cyprus.

Abstract. Cultural assets of historical value need to be holistically documented to ensure their long-term preservation. A key component of the holistic documentation of a cultural heritage object is the materials' characterization by means of analytical techniques. In this work we have employed non-invasive analytical techniques (x-ray fluorescence, Raman spectroscopy and computed tomography) for the study of three ceramic zoomorphic rhyta, belonging to the museum of George and Nefeli Giabra Pierides located at the Bank of Cyprus Cultural Foundation in Nicosia, Cyprus. The plain ware rhyta in the form of a he-goat are dating to the Hellenistic period (310-30 B.C). We have used non-invasive portable XRF and Raman spectroscopy for the in situ chemical characterization of color traces preserved at the surface of the three zoomorphic rhyta. In addition, we have employed Computed Tomography (CT) as a non-invasive method to investigate the manufacturing technique of the ceramics. CT was used to visualize manufacturing details and to provide the accurate profiles of the ceramics, especially of the closed shapes. CT enabled a closer examination of the objects and generated a three-dimensional (3D) digital model of the artefacts which was used to replicate them using additive manufacturing (3D printing).

Keywords: ceramics, cultural heritage, computed tomography, 3D printing, XRF, Raman spectroscopy.

1 Introduction

The archaeological collection of George and Nefeli Tziapra Pierides of the Bank of Cyprus Cultural Foundation, gathers among others, animal-shaped vases of the Early and Middle Bronze Age (Karageorghis & Boardman, 2002), which are unique and demonstrate the inventive spirit and sense of humour of the Cypriot artist of this period.

One represents a ram and the other probably a donkey. According to researchers, these are funerary offerings, probably from Larnaca cemeteries. These grave goods accompanied the deceased, who may have been a farmer, to his last residence (Karageorghis & Boardman, 2002). A horse mask, unique of its kind, belonging to the Tziapra Pierides Collection, is dated to the Cyproarchaean I-II period. It should also be noted that the clay animal masks that have survived are generally bull masks.

In this work, we have employed non-invasive analytical techniques for the study of three ceramic zoomorphic rhyta, belonging to the museum of George and Nefeli Giabra Pierides located at the Bank of Cyprus Cultural Foundation in Nicosia, Cyprus (Figure 1) (Karageorghis & Boardman, 2002). The Hellenistic pottery that has been found proves that Cyprus has an important tradition in ceramics, along with the large quantities of imported pottery that it receives from all over the Mediterranean. The three animal-shaped rhyta of simple goat form (with inventory codes 241, 242, 243)(Karageorghis & Boardman, 2002), which are prominently exhibited in the museum of the George and Nefeli Tziapra Pierides collection, are also part of this context (Karageorghis & Boardman, 2002). The animals are rendered with pragmatic features, especially their heads. Archaeologists suggested that the three plain ware rhyta have been used as 'feeding bottles'.

The heads of goats are compact, have twisted horns and two of them have beards. They have elegantly constructed ears, eyes and muzzle. They have spots on the forehead. The body is cylindrical and wheeled and rests on four short legs. The animals have a short tail and a reed-shaped handle at the back of the neck. The sternum has a short and narrow forechest. There is a filling hole at the back of the handle. These three rhyta come from the same pottery workshop and probably from the same craftsman(Karageorghis & Boardman, 2002).

Figure 1: Three plain ware rhyta in the form of a he-goat. Hellenistic period 310-30 B.C. Style: Plain ware. Material: Clay Source: Photographs reproduced from the Bank of Cyprus Cultural Foundation.

The preservation of tangible cultural heritage and its intangible information requires a holistic documentation approach. The development of such approaches requires a collaborative and multidisciplinary research, including the three-dimensional documentation of cultural heritage assets (data acquisition, data processing and modelling), the materials analysis of the artefacts, the knowledge management, and the use and re-use of the acquired information (Ioannides & Davies, 2019). Cultural assets of historical value need to be holistically documented to ensure long-term preservation, to define authenticity, to prevent or identify illicit trafficking and to assist cultural heritage storytelling via new technologies, e.g., augmented reality (AR) and virtual reality (VR) applications.

A key component of the holistic documentation of a cultural heritage object is the materials' characterization by means of analytical techniques. Natural sciences have provided valuable tools and strategies for the analytical research of cultural objects and opened up new ways to diagnose, monitor and protect them. The scientific analysis of cultural heritage artefacts can provide information on the manufacturing technique, elucidate chemical components attributed to the nature and/or manufacture of materials, or to subsequent restoration processes. These material analyses may include scanning electron microscope and energy dispersive X-ray analysis (Schreiner et al., 2007), Fourier transform infrared (FTIR) spectroscopy and microscopy (Prati et al., 2010), Raman spectroscopy (Casadio et al., 2017), X-ray fluorescence (XRF) spectroscopy (Janssens et al., 2000), X-ray diffraction (XRD) (Gonzalez et al., 2020), ultraviolet-visible-near infrared (UV-Vis-NIR) absorption and emission (Picollo et al., 2019), hyperspectral imaging (Picollo et al., 2020), x-ray computed tomography (CT) (Morigi et al., 2010), immunochemical methods (Cartechini et al., 2017), gas chromatography mass spectrometry (GC-MS) (Bonaduce et al., 2017), high performance liquid chromatography (HPLC) (Degano & la Nasa, 2017), radiocarbon dating (Hajdas et al., 2021) and synchrotron based techniques (Bertrand et al., 2012).

We have used non-invasive portable XRF and Raman spectroscopy for the in situ chemical characterization of color traces preserved at the surface of the three zoomorphic rhyta. XRF allows a rapid identification of the elemental composition of a material. It is a primary tool commonly employed as a first approach of an in-situ study. In situ Raman spectroscopy is a relatively quick analysis applied directly at the surface of the artefact that can be employed for the identification of both organic and inorganic molecules. Raman spectroscopy provides a molecular fingerprint of the compounds under study and can be used to identify the materials of artworks and archaeological artefacts or to elucidate their conservation state (Rousaki & Vandenabeele, 2021). In this study, coupling the two techniques provided information on the elemental and molecular composition of the ceramics' matrices and pigments used.

In addition, we have employed CT as a non-invasive method to investigate the manufacturing technique of the ceramics. CT was used to visualize manufacturing details and to provide the accurate profiles of the ceramics, especially of the closed shapes. CT enabled a closer examination of the objects and generated a three-dimensional (3D) digital model of the artefacts which was used to replicate them using additive manufacturing (3D printing).

2 Materials and methods

2.1 X-ray Fluorescence spectroscopy

For the XRF measurements we have used the Thermo Scientific portable XRF Niton XL5 spectrometer (Figure 2). The analytical range was from Mg to U, the X-Ray Tube was Ag anode and the spot size was 8mm. XRF Spectra graphs analyses were carried out with the use of the scientific software Thermo Scientific NDT.

Figure 2: The portable XRF Niton XL5 spectrometer.

2.2 Raman Spectroscopy

For the Raman spectroscopy measurements, we have employed the portable DeltaNu RockHound with 785 nm laser source, the resolution was 8 cm-1 and the spectral range was from 200 to 2000 cm-1 (Figure 3). Raman spectra graphs analyses were carried out with the use of the scientific software Spectragryph 1.2. The identification of pigments for Raman Spectroscopy was done using the UCL (Clark) data base, Pigments Checker and colourlex.com for pigment analyses of paintings.

Before the main process of pigments identification, Raman spectra were subjected to a pre-processing procedure such as baseline correction, Savitzky - Golay smoothing and normalization.

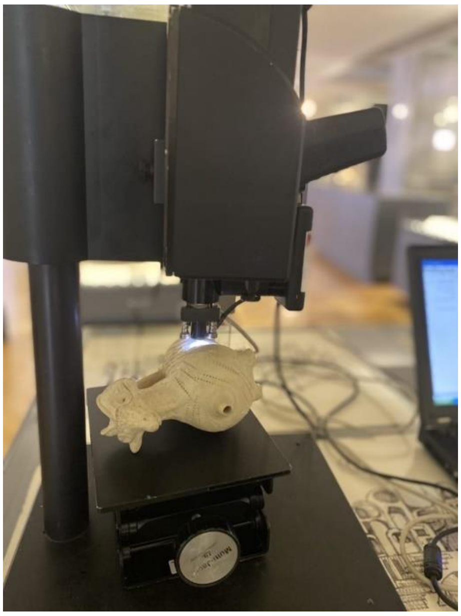

Figure 3: The portable DeltaNu RockHound Raman spectrometer.

2.3 X-ray Computed Tomography

One of the main objectives of the use of the 3D X-Ray Computed Tomography Systems, is to provide a high-quality visual documentation. CT has significant accuracy in capturing geometry and delivers a highly accurate 3D mesh in a high resolution, gives clearly visible of the outer surface, through the CT surface reconstruction.

Additionally, important is its ability to investigate a closed vessel or within the ceramic material itself. Structure and pores in the ceramics can be analysed according to amount, size, shape, and orientation. CT shows and unveils significant details of the manufacturing process of the artefacts. For detecting the incisions, we provided high resolution geometric data with the usage of appropriate small measuring fields.

The main challenge for CT experiments was the transfer of precious samples from the Bank of Cyprus Cultural Foundation museum to the CT laboratory of Ygeia Polyclinic. The three ceramic zoomorphic rhyta were received accompanied by a representative from the museum and with attention one by one to were placed on the examination scan bed.

The experiments were performed with a multidetector CT scanner with 128 slices per rotation without administration of any material. Scanning was performed with a slice thickness of 0.625 mm, a 0.4 ms lamp rotation and FVO 512×512 cm^2. Volumes were reconstructed in solid and soft window but mainly with grey levels imaging counting

Hounsfield values to record the density of its wall's construction material in various parts of the inner surface.

We reconstructed 3D digital models of the artefacts which aid on the detailed analysis on both the exterior and interior of the statuettes, comparing among themselves, in terms of shape, spatial volume, joints, as well as some damages and cracks.

2.4 3D reconstruction and replication using additive manufacturing (3D printing)

The image stack from the CT scans was processed to produce a 3D shell surface. The surface was transformed into a volumetric structure. The 3D solid was then verified for any discontinuities and improvements were taken place. Appropriate dimensions were applied through scaling and the geometry was prepared in terms of printing direction and temporary supports, for 3D printing using an SLA resin printer (Figure 4). A Formlabs Form3L 3D printer of 0.025 mm transverse resolution was used with white resin V4 material set for 0.1 mm layer thickness. The manufacturing process lasted approximately 27h for 1301 layers using a total of 289.66 ml of resin.

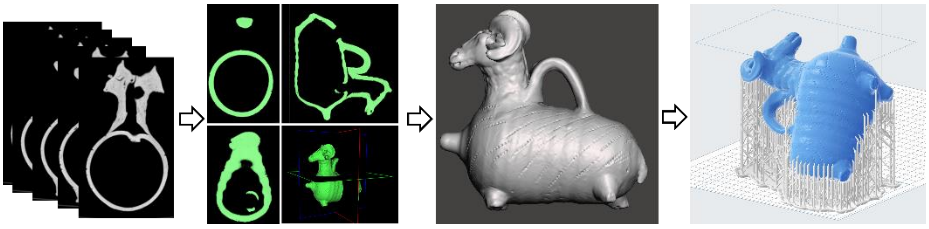

Figure 4: *Schematic representation of the process from CT scans to 3D printing.*

3 Results

3.1 Holistic Documentation of Zoomorphic Rhyta

The conceptual model of data presented in Figure 5 aims on the holistic documentation of the zoomorphic rhyta and divides the types of data needed for the record of the cultural heritage objects into classification categories. The taxonomical system was created under the MNEMOSYNE project (H2020 ERA Chair) categorizes the zoomorphic rhyta into the classes of "Tangible", "Movable", "Function", "Furnishings and Equipment", "Containers", "Culinary Items", "Drinking vessels", "Rhyta" (Figure 5). The intangible information of the objects is recorded in the classes "Context" and "State/Condition" (Figure 6, Figure 7). Furthermore, we represent the data needed to record the tangible facet of the object in classes such as "Form" and "Technique" (Figure 6, Figure 11).

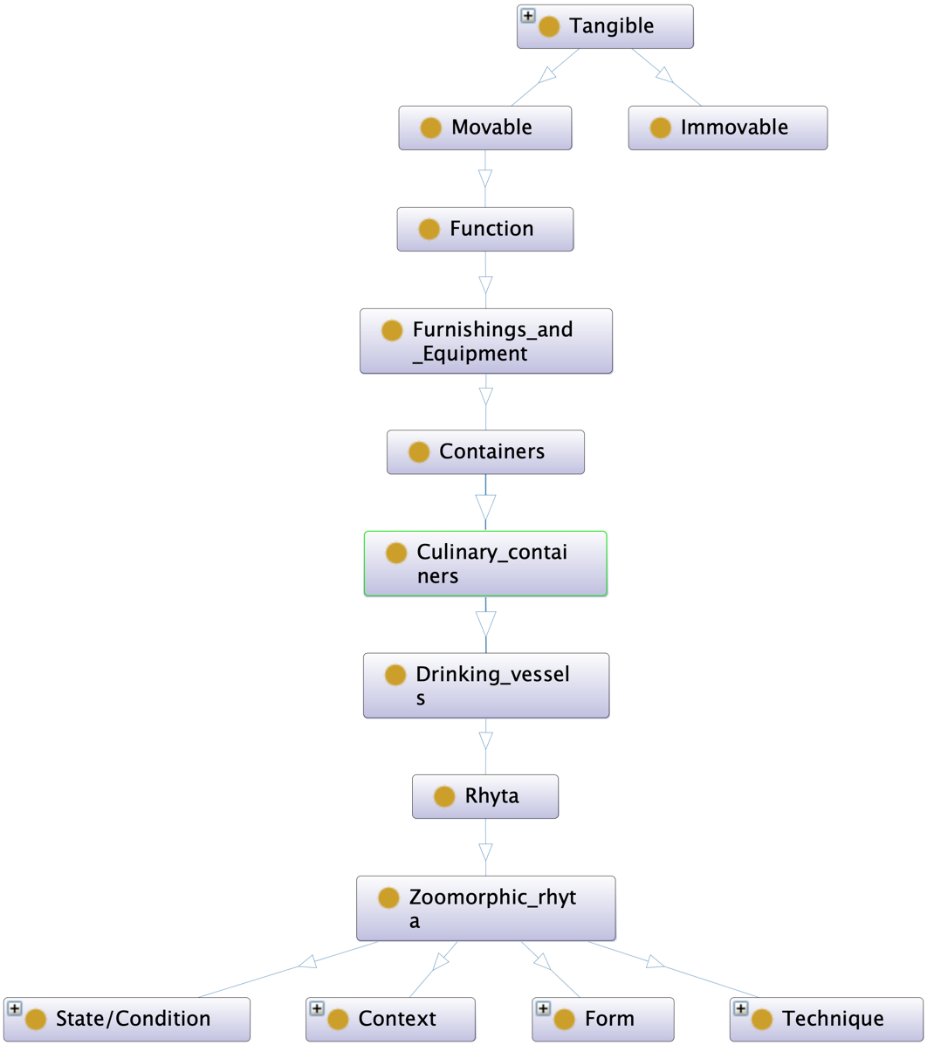

Figure 5: Conceptualization of holistic documentation regarding the Zoomorphic Rhyta.

Within the “Context”, the data is separated into “Location” and “Period” which includes information about the original ancient production of the object. Within “State/Condition”, the data is separated into the “Post-depositional” information regarding the zoomorphic rhyta, which includes information on the collection in which the artefacts belong and the “Pre-depositional” information which are unknown (Figure 7, Figure 10, Figure 9, Figure 10). This data offers important information regarding the history of the objects. The other categories, “Form” and “Technique” provide information related to the tangible facet of the physical objects (Figure 6, Figure 11, Figure 12, Figure 13, Figure 14, Figure 15).

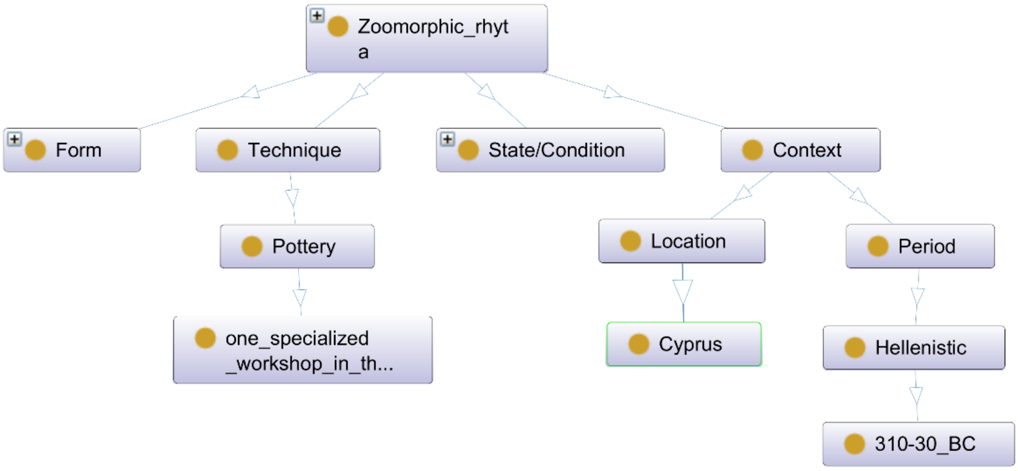

Figure 6: "Context" and "Technique" taxonomy.

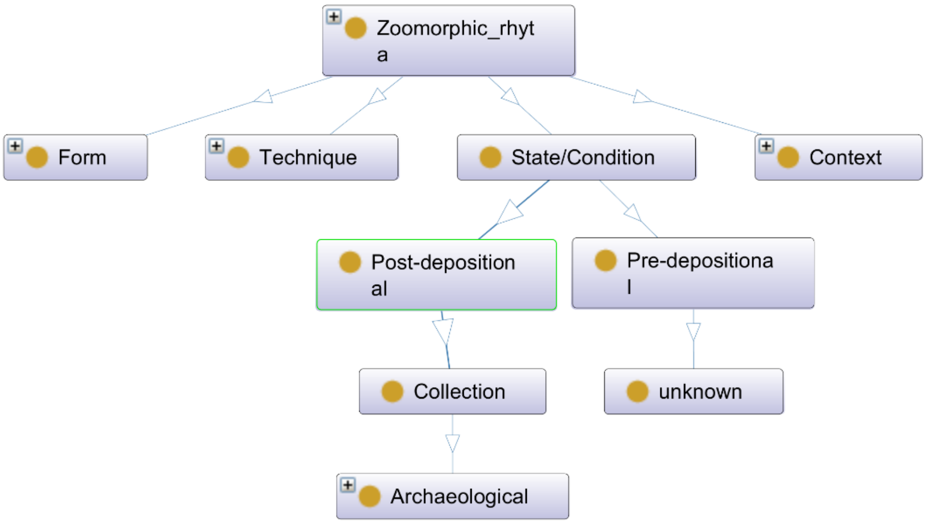

Figure 7: "State/Condition" taxonomy. The expansion of the "Archaeological" branch is shown in Figure 10.

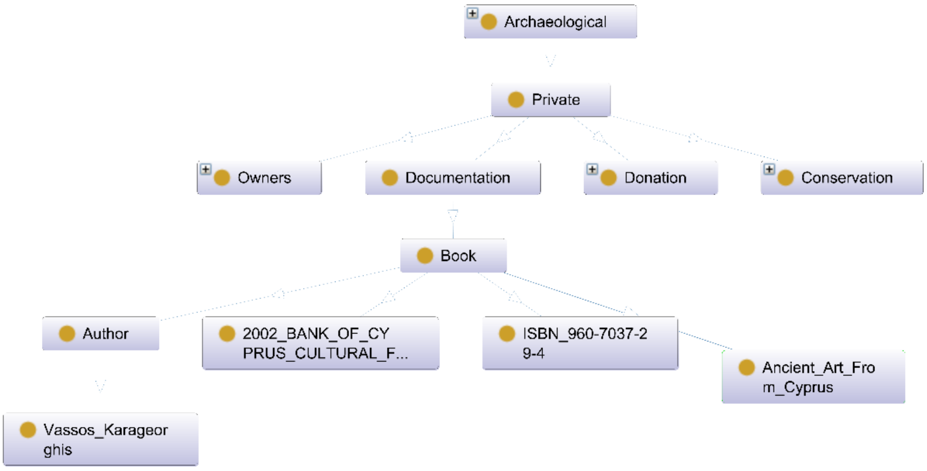

Figure 8: "Collection" taxonomy: Expansion of the "Archaeological branch" (1).

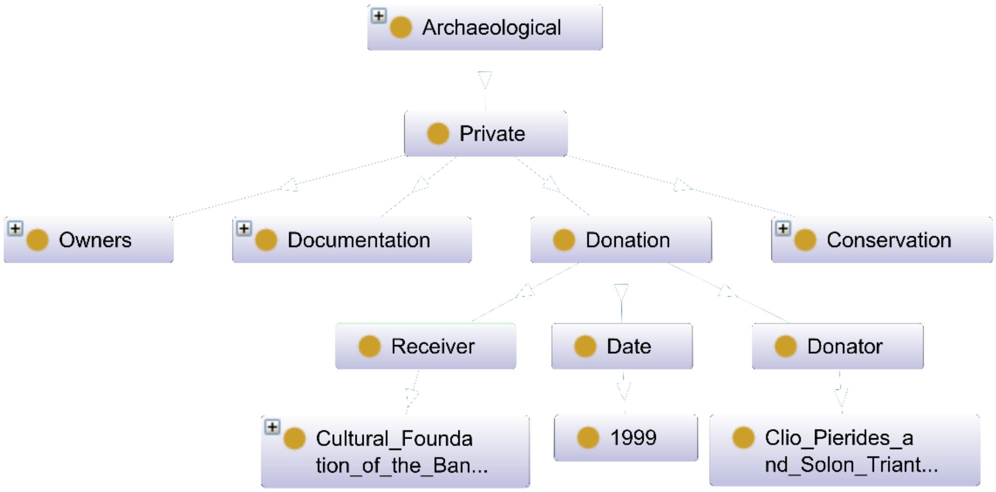

Figure 9: "Collection" taxonomy: Expansion of the "Archaeological branch" (2).

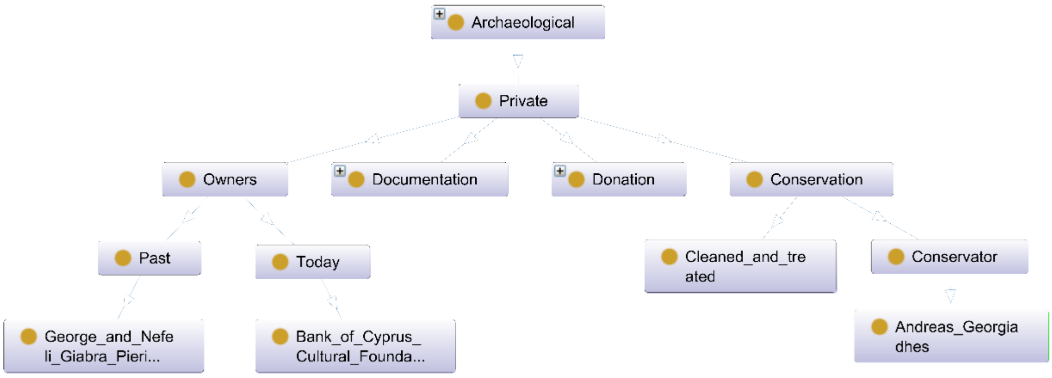

Figure 10: "Collection" taxonomy: Expansion of the "Archaeological branch3" (3).

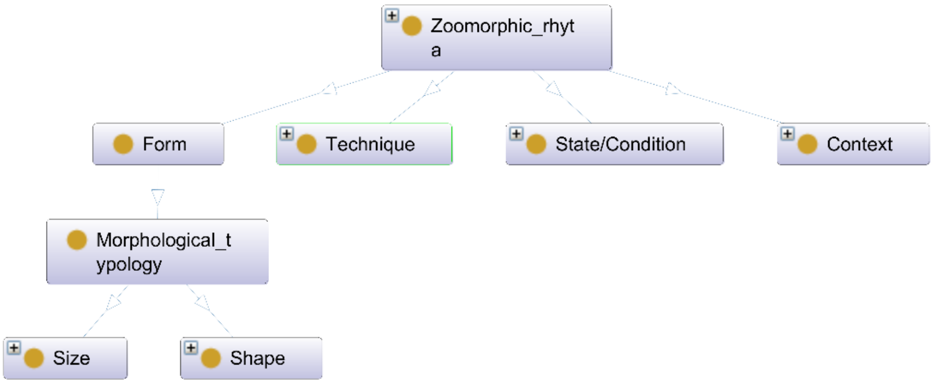

Figure 11: "Form" taxonomy. The expansion of the "Morphological typology" branch is shown in Figure 12 and Figure 13.

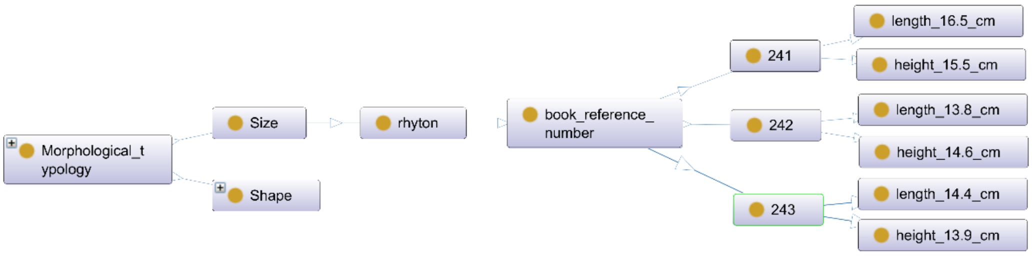

Figure 12: "Form" taxonomy: Expansion of the "Morphological Typology-Size" branch.

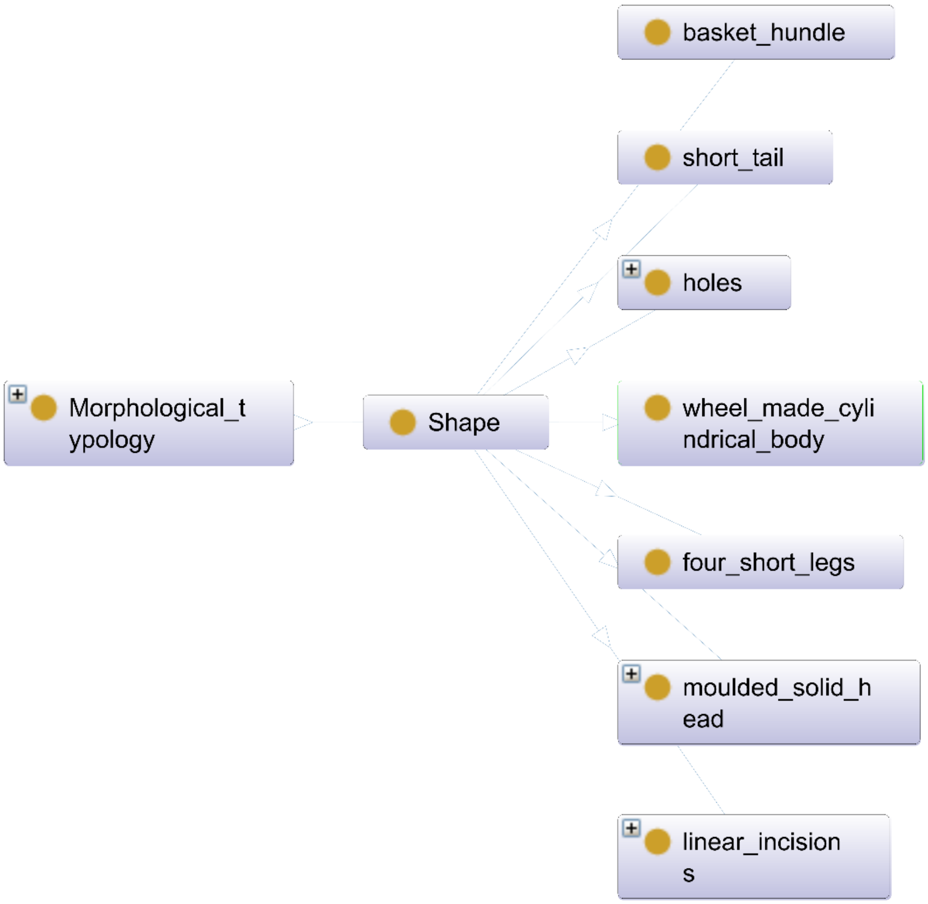

Figure 13: "Form" taxonomy: Expansion of the "Morphological Typology - Shape" branch (1).

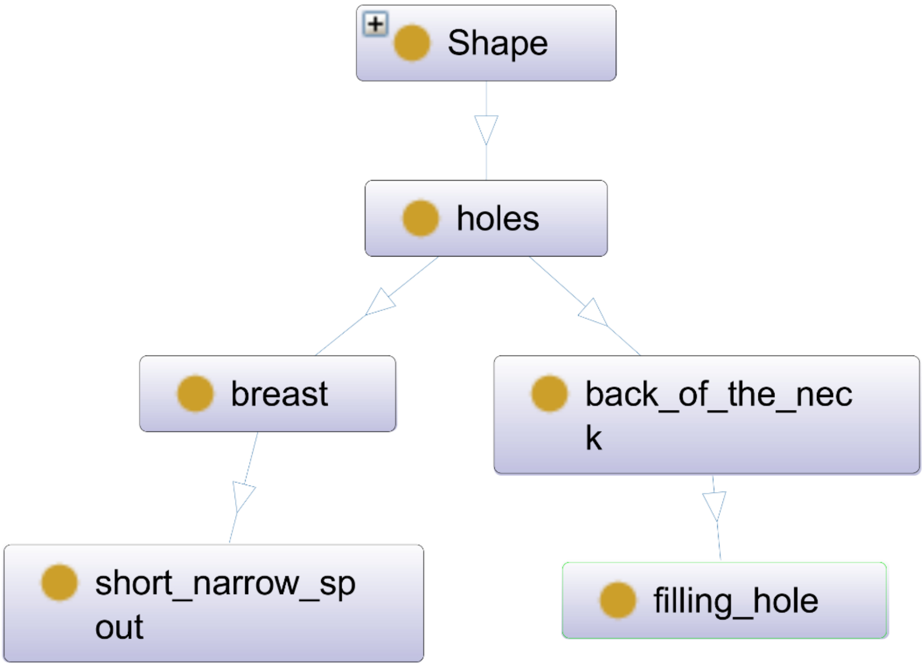

Figure 14: "Form" taxonomy: Expansion of the "Morphological Typology - Shape" branch (2).

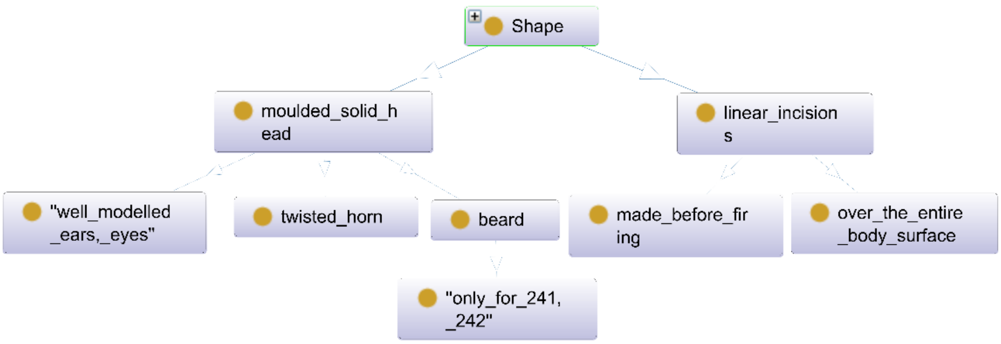

Figure 15: "Form" taxonomy: Expansion of the "Morphological Typology - Shape" branch (3).

3.2 Pigments Identification

The development of new methods and data treatment of the spectral information is a field of continuous research in the field of Cultural Heritage where real samples are always complex mixtures of original and degradations compounds that require new approach to be implemented in the daily practice of Raman spectroscopy (Christopoulou et al., 2021; Ganetsos et al., 2020; Lukačević et al., 2016).

The presence of manganese black was identified in all zoomorphic rhyta. The ceramic decoration techniques in Cyprus were investigated by (Aloupi et al., 2000). From the end of the Late Bronze Age (1050 - 325 BC) onwards, Mn ores (umbrae) were used for the production of dark colours. Mn ores have varying Mn_3O_4 (2.5-15 %) and Fe_2O_3 (20-65 %) contents. The pigment is prepared easily with firing in kiln atmosphere. (*Figure 16*) (Manganese Black - CAMEO). The pigment is stable in acids and alkalis and also at high temperatures, shows excellent lightfastness, and is compatible with all other pigments.

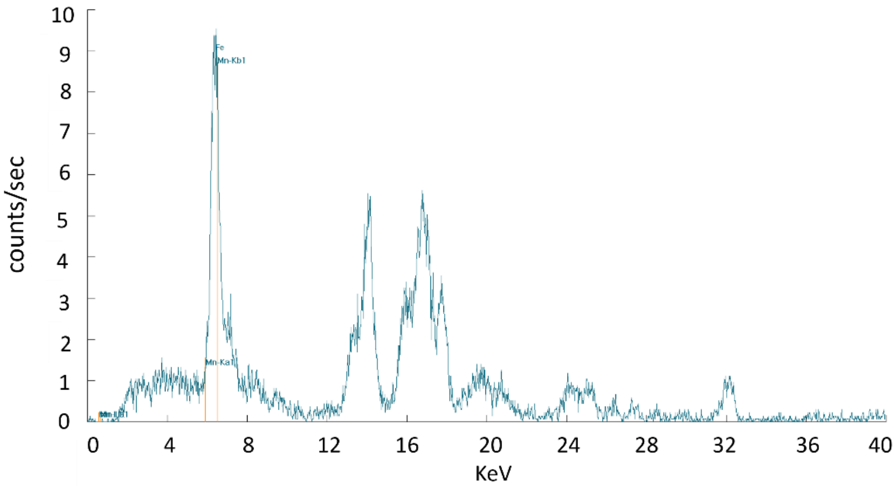

Figure 16: XRF spectrum collected from artefact GC21.

In addition, we identified red ochre (Figure 17). The main color giving component of natural red ochre (ocher) is composed of hematite ($\propto$-Fe_2O_3). The mineral is abundant in Cyprus (Aloupi et al., 2000) and in past studies red ochre was identified in wall paintings of monasteries (Daniilia et al., 2008). Iron oxides are stable at high temperatures but not resistant against acids. The pigment is absolutely stable as is documented by the cave paintings still in excellent condition after many thousands of years. We defined red ochre in all artefacts (Aloupi et al., 2000; Gasanova et al., 2018).

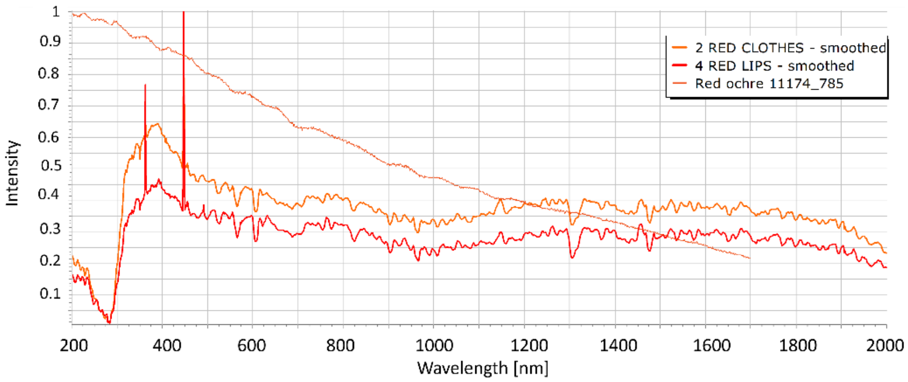

Figure 17: Raman spectrum collected from artefact GC28 compared with red ochre reference sample (785 nm).

We present the final results of each technique and also the pigment identified using both results in the Table 1 (Caggiani et al., 2016).

Table 1. Identified pigments as a result of the complementary XRF and Raman characterization

Artefact	Colour Hue	XRF (elements)	Raman peaks (cm^{-1})	Pigment identified
GC21, GC22, GC28	**Black**	*Ca, Fe, Mn*	961m; ~ 1325vs; ~ 1580vs	Carbon black and iron-manganese
GC21, GC22, GC28	**Red**	*Fe, Ca*	220vs; 286vs; 402m; 491w; 601w	Red ochre and Hematite (Fe_2O_3 + clay + silica)

3.3 X-ray Computed Tomography

CT was employed to visualize the closed shape structures of the museum objects and to provide details on the manufacturing technique of the three ceramics. CT data is recorded in grey levels due to the local material density (Karl et al., 2014). Inaccessible parts of the objects became visible and can be therefore evaluated to decipher the technique behind their manufacture (Bouzakis et al., 2011). The two-dimensional virtual cross sections of the objects show that molds were used to create the left- and right-side of the head (see arrows Figure 18), which were connected to the body at a second stage (Figure 18). The characteristic helical structure of the body suggests that it was made using a wheel (Figure 19); the helical structure derives from the craftsman fingers creating the shape of the ceramic (Bouzakis et al., 2011). The 2D virtual cross sections obtained with CT illustrates the differences in the handcrafted making of the openings (Figure 20). CT allows the 3D visualization of closed shaped museum objects, the collection of volumetric data and the calculation of the ceramics volume and provides insights of the manufacturing technique, as well as materials characterization.

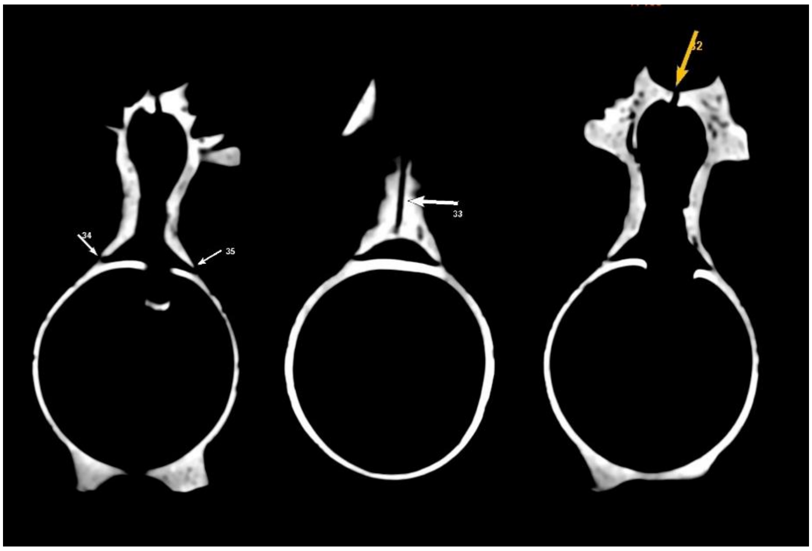

Figure 18: CT data. Two dimensional virtual cross sections of the zoomorphic rhyton GC21 showing the manufacturing details of the head.

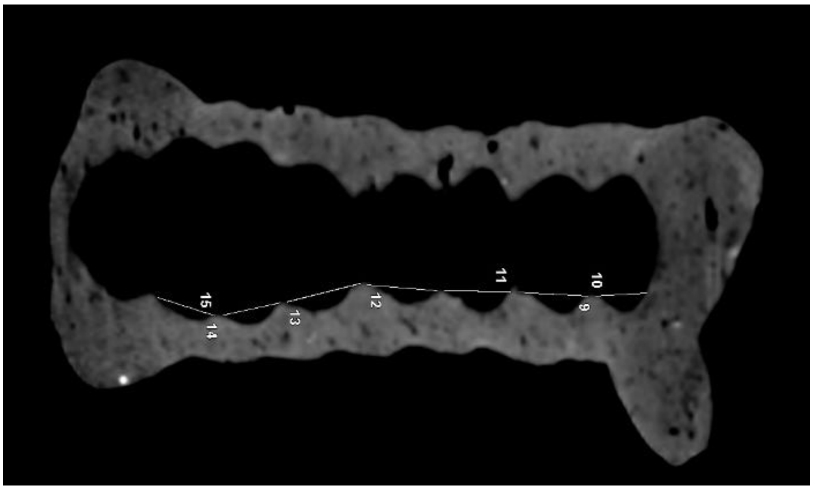

Figure 19: CT data. Two-dimensional virtual cross sections of the zoomorphic rhyton GC21 showing a characteristic helical structure of the inner body.

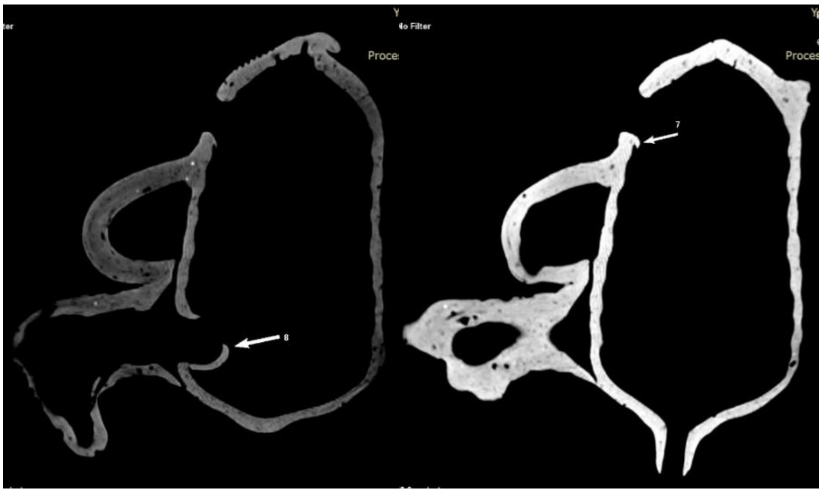

Figure 20: CT data. Two-dimensional virtual cross sections of the zoomorphic rhyton GC21 showing the openings of the ceramic.

The manufacturing technique of the zoomorphic rhyton GC28 is distinguishable. The 2D virtual cross sections of the artefact GC28 show that two types of clays have been used for its manufacture. The two different grey colors displayed in Figure 21 correspond to materials of different chemical composition.

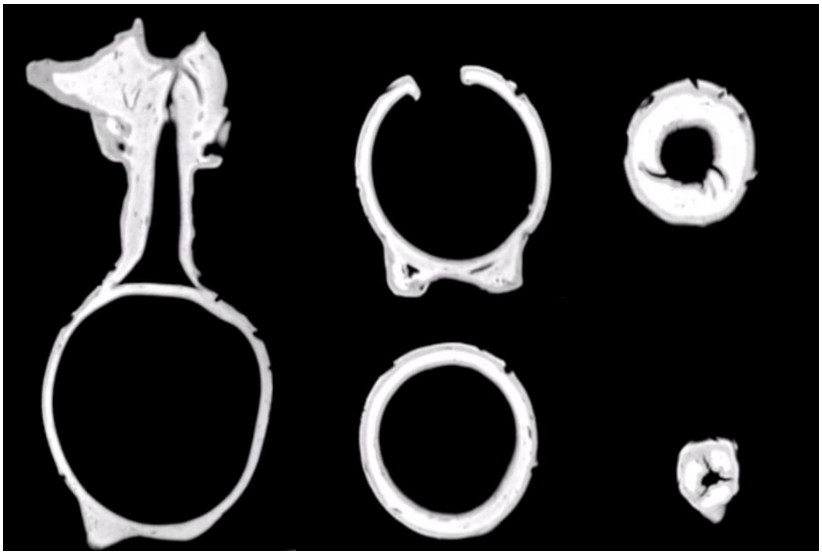

Figure 21: CT data of zoomorphic rhyton GC28. The two grey levels present in the 2D sections of the artefact show that two types of clays have been used for its manufacture.

3.4 3D reconstruction and replication using additive manufacturing (3D printing)

Figure 22 shows the 3D reconstructed solids and in a semitransparent view in which the wall size and inner cavity are visible. The difference in the head cavity between the two artifacts can be observed. The rhyta are of similar style and decoration. Superimposing the two rhyta i.e. C21 and C28, sizes can be compared (Figure 23).

The chosen 3D printing method, material and equipment seem to have sufficiently captured the texture and decorative elements of the zoomorphic rhyton (Figure 24). In replicating artifacts using additive manufacturing methods, several factors might need to be considered and optimized for the specifics of the artifact. Such factors are the 3D printing method, e.g., SLA resin based or FDM, the material, specifics of the geometry, the resolution required to capture details of relevance and archeological importance for the artifact, etc. In this study, the orientation of the specific object during printing and the geometry of the artifact (related to its function) can result in cups, i.e., regions where liquid resin might concentrate adding to the weight of the material and causing collapse or introducing cracks. Mitigating strategies might involve hollowing the walls in order to reduce weight (and amount of used material) and introducing channels for the excess resin to escape the cup regions. However, any added features for the purposes of manufacturing should be considered in relation to the purpose of replication so as to avoid alterations to the artifact that relate to its purpose and thus rendering the replication ineffective (e.g. adding channels could potentially hinder the study of liquid flow through the replicated rhyton and introduce holes on the exterior surface texture).

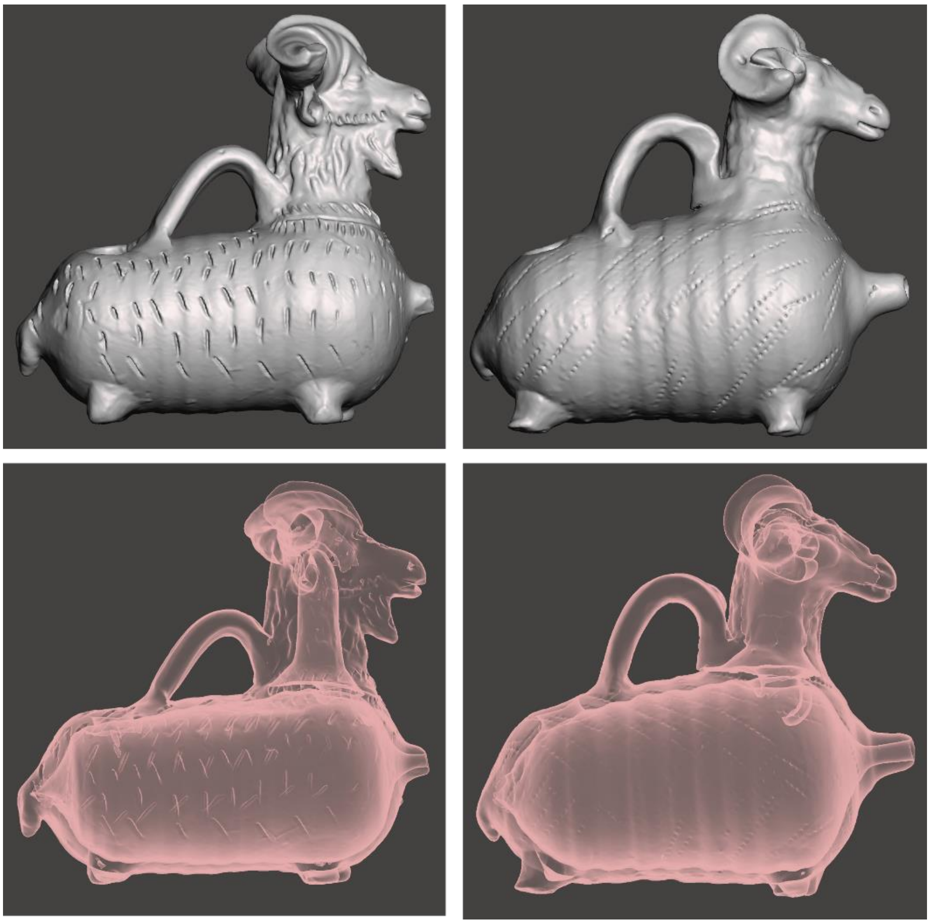

Figure 22: 3D reconstructed rhyta with exterior decoration (top), semitransparent view of the artifacts in which wall size and cavity are visible.

Figure 23: Comparison of the size of rhyta C21 and C28 by superimposing the reconstructed volumes.

Figure 24: 3*D printed replica of zoomorphic rhyton from CT scans.*

3.5 Future work

Archaeologists suggest that the three plain ware rhyta in the form of a he-goat have been used as ‘feeding bottles’, however, there is no analytical data to support the claim. Biomolecular components of organic materials associated with human activity may have survived on the inner part of the potteries. Future work will aim on the identification of the nature and origins of any organic residues present using analytical organic chemical techniques that can reveal the principal use of the rhyta.

The application of separation (chromatographic) and identification (mass spectroscopic) techniques can reveal preserved and altered biomolecular components of organic residues that hold archaeological information. In this way we can answer the long-held archaeological hypothesis and offer a new perspective on the study of human activity in the past.

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