ISIS@MACH ITALIA

Science and Technology Facilities Council
ISIS Neutron and Muon Source

Experiment Proposal

Experiment number GP2023091

Principal investigator	Dr Giulia Marcucci, ISIS Neutron and Muon Source, UNITED_KINGDOM
Co-investigator (*)	Dr Daniela Di Martino, University of Milano Bicocca, ITALY
Co-investigator	Dr Massimiliano Clemenza, INFN, ITALY
Co-investigator	
Co-investigator	
Co-investigator	
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Co-investigator	
Co-investigator	
Experiment title	Unlocking the structure and composition of a historical silver coin using Wide Angle X-ray Diffraction in combination with Muon and Neutron Techniques
MRF Instrument	**SAXS WAXD**
Access Route	Direct Access
Science Areas	Cultural Heritage, Materials
Sponsored Grant	None
Grant Title	-
Start Date	-
Similar Submission?	-
Industrial Links	-

Days requested: 1
Previous GP Number: NO
DOI: -
Sponsor: -
Grant Number: -
Finish Date: -

Non-Technical Abstract

The INFN has funded the CHNET_TANDEM collaboration aimed at the development of a non-destructive analytical technique using negative muon beams. As part of this effort, an 18th-century Portuguese coin was used to compare the muon technique with other methods, in collaboration with the IAEA. The muon beam technique revealed the coin elemental composition and depth profile, showing a possible silver enrichment. The main objectives of this proposal are to further investigate the coin composition and structure with a non-destructive approach exploiting the complementarity of information of a multi-technique protocol. We plan to use Wide Angle X-ray Diffraction to investigate the structure and potential alterations in the coin phases structure and help understand its production technology, in combination with XRD tomography and cutting-edge scientific techniques with heritage science to assess the results obtained with previous muons and neutrons analyses.

Publications -

ISIS neutron and muon source

E-platform: No

Instruments	**Days Requested:**
Access Route	**Previous RB Number:**
Science Areas	**DOI:**
Sponsored Grant	**Sponsor:**
Grant Title	**Grant Number:**
Start Date	**Finish Date:**
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Sample record sheet

Principal contact Dr Daniela Di Martino, University of Milano Bicocca, ITALY
MRF Instrument **SAXS WAXD** **Days Requested:** 1
Special requirements:

SAMPLE

Material	Copper and Silver coin	-	-
Formula	Cu, Ag	-	-
Forms	Solid		
Volume	0.22 cc		
Weight	2 g		
Container or substrate	no	-	-
Storage Requirements	-	-	-

SAMPLE ENVIROMENT

Temperature Range	room temperature - K	-	-
Pressure Range	no applied pressure - mbar	-	-
Magnetic field range	no applied magnetic field - T	-	-
Standard equipment	None	-	-
Special equipment	none	-	-

SAFETY

Prep lab needed	No	-	-
Sample Prep Hazards	no	-	-
Special equip. reqs	none	-	-
Sensitivity to air	No	-	-
Sensitivity to vapour	No	-	-
Experiment Hazards	no	-	-
Equipment Hazards	-	-	-
Biological hazards	no	-	-
Radioactive Hazards	no	-	-
Additional Hazards	-	-	-
Additional Details	-	-	-
Sample will be	Disposed by IS	-	-

Background and Context

The INFN has funded the CHNET_TANDEM collaboration aimed at the development of a non-destructive analytical technique for Cultural Heritage using negative muon beams. Proof-of-principle experiments using negative muons for elemental analysis were conducted on the Port 4 beamline of the ISIS Neutron and Muon Source from April 2015, including calibration on standard materials [1] and feasibility tests on at many archaeological artefacts, such as “bronze age” artefacts (CHNET_TANDEM INFN experiment), Roman Empire coins and ancient swords to name but a few [2-4].

As part of this project, an 18th-century Portuguese coin has been used for a round-robin comparison in participation to the IAEA (International Atomic Energy Agency) Coordinated Research Project (CRP) F11021 [5] “Enhancing Nuclear Analytical Techniques to Meet the Needs of Forensic Science” with the Muonic Atom X-ray Spectroscopy performed at PORT4 of the ISIS Neutron and Muon Source. This CRP allowed introducing, in the IAEA framework, the use of negative muons as a reference technique for non-destructive elementary characterization measurements for unique samples, such as those of cultural heritage or those measured for forensic reasons.

The application of the Muonic Atom X-ray spectroscopy allowed to perform an elemental depth profile of the coin, determining the Ag/Cu ratio from the surface to the inner core of the sample and therefore disclosing a slight silver enrichment, as shown in Fig. 1. Preliminary XRF measurements were carried out and main results for composition are listed as follow: Ag: 91.2%, Cu: 3.7%, Cl: 1.2% Au: =0.7%, Fe: 0.5%, Pb: 0.2% plus other minor components. We can also confirm that, on the surface, the coin is silver-based, with copper as a minor alloy constituent and other elements between 0.2 -1%. The main interest of this proposal is to cross-check this relatively new nuclear investigation with consolidated non-invasive techniques to reveal the exact composition (surface and bulk) and homogeneity along the depth profile and to expand the punctual elemental analysis to the phase composition representative of the entire sample, to also determine the production process, whether by minting or casting.

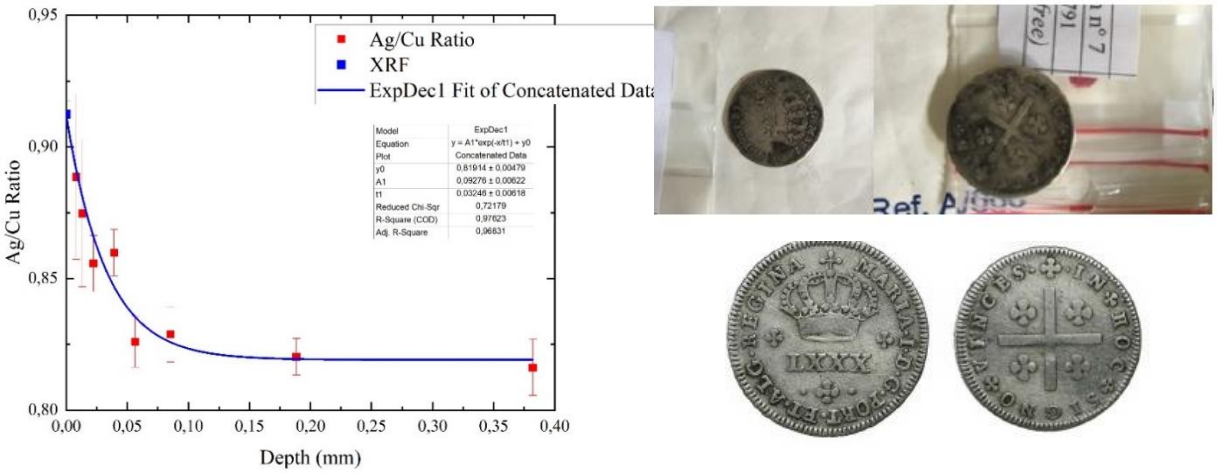

Fig. 1 On the left: Depth profile of the Ag/Cu ratio obtained through Muonic Atom X-ray spectroscopy measurements at the ISIS Neutron and Muon Source. On the right: (top) Front and rear of the Portuguese coins, 80 reis, coinage under Maria I (2 cm in diameter and 0.7 mm in thickness). (bottom) A Portuguese coin, 80 reis (coinage under Maria I) from a recent auction [6].

Sample description

A Portuguese coin, dating to the late 18th century will be investigated and is part of the round-robin comparison in CRP F11021. This coin is shown in Fig. 1. During the 18th century, the Portuguese monetary unit was the reis. The etymology comes from “*rei*” (literally meaning king), the plural being “*reis*”. Different types of coinage can be found and are either copper-, silver- or

gold-based. The Portuguese coinage consisted of 5, 10, 20 and 40 reis pieces in either copper or bronze; a silver coinage of 60, 80, 120, 200 and 400 reis and gold coinage of 480, 800, 1,200, 1,600, 3,200 and 6,400 reis. Our coin has inscribed on it "*LXXX*" and is therefore 80 reis. In addition, the name of the queen (Queen Maria I who ruled from 1777 to 1799). A picture of the sample (front and rear) is shown below.

Proposed experiment

The primary objectives of this study are as follows:

i. Phase composition and distribution analysis: Perform XRD tomography to determine the precise composition of the coin, including the ratio of silver to copper and the presence of any alloying elements to cross-check the Muonic Atom X-ray Spectroscopy results;
ii. Structural Composition: Investigate the structure and potential alterations in the coin's phases structure by WAXD analysis caused by historical factors such as copper depletion and minting techniques.
iii. Historical Context: Correlate the findings with historical records and numismatic data to provide insights into the coin's origin, purpose, and significance.

We propose to use WAXD analysis to accomplish our research objectives, considering also this three-fold motivation: 1) the sample is an ancient artefact, and non-destructive analyses should be used to preserve its uniqueness; 2) no cleaning will be performed on the sample– we will be able to perform the measurement also in the presence of corrosion layers or deposits, suggested by XRF measurements; 3) the sample is bulky, and we want to infer not only the mean bulk composition but the depth profile. In this regard, another proposal will be submitted for the same sample for XRD tomography measurements to accomplish the phase composition investigation. These two experiments will be useful in complementing the information collected through neutron diffraction and neutron resonance capture analysis carried out at the INES beamline at ISIS (RB2010534, "Combination of neutron based techniques to derive the composition of an 18th-century coin")."

We would like to underline that this round-robin is on a real sample. Other measurements have been done on standards; however, the study of a real case is mandatory when these techniques are to be used on real specimens and historical artefacts are always not homogeneous and present different issues in comparison to a standard sample.

Therefore, we aim to measure n. 1 sample in three different positions with a Cu Kα radiation source, in the diffraction range up to 60° 2theta. Hence, we request, I day which accounts also for setup time.

References

[1] A.D. Hillier et al, Microchemical Journal. Vol. 125, March 2016, Pages 203–207.
[2] M. Clemenza et al. Nucl. Instrum. Meth. Phys. Res. A . 936, (2019), Pages 27-28
[3] A.D. Hillier, A. M. Pollard, A. Wilson, D. MckPaul, et al in prep, see expt report RB 1520462.
[4] A. I. Wilson 'The metal supply of the Roman Empire', in E. Papi and B. Scardigli (eds), Supplying Rome.
[5]A. Fajgelj et al The IAEA's Analytical Quality Control Services (AQCS) Programme on Intercomparison Runs and Reference Materials. IAEASM-344/3. 1997
[6] See for example "Lot 337 Auction 23" where 4 of these coins were estimated at 40 euros https://numismaticaleiloes.bidinside.com/en/lot/335/portugal-d-pedro-ii-to-d-maria-i-4-/