NEUTRON TECHNIQUES IN CULTURAL HERITAGE

G. FESTA 1, C. ANDREANI 1, A. FILABOZZI 1, D. MALFITANA 2, J. POBLOME 3

1 Università degli Studi di Roma Tor Vergata, Department of Physics, Via R. Scientifica 1, 00133, Rome, Italy
2 CNR – Istituto per i Beni Archeologici e Monumentali, IBAM Catania, Italy
3 Katholieke Universiteit Leuven (Belgium)

Kivonat

A szerzők római kori márványtöredékeket vizsgáltak a Villa Adriana anyagából (Tivoli, Róma) neutron diffrakciós módszerrel. A vizsgálatok célja az volt, hogy az ásványfázis összetételét és a szöveti jellemzők lapján el tudják különíteni a különböző márványtípusokat. A szöveti jellemzők, egyéb szerkezeti részekekek együtt, felhasználhatóak a márvány, kerámia és bronz anyagú régészeti leletek származási helyének megállapítására és segíthetek azok szakszerű konzerválását is.

Abstract

Roman marble fragments from the Villa Adriana at Tivoli (Rome) have been characterised by neutron diffraction. This study aimed at distinguishing between different marble types on the basis of the mineral phase compositions and the crystallographic textures. The texture information, along with other structure details, can be used as characterising feature to address questions of attribution and restoration of archaeological marble, ceramic or bronze objects.

KULCSSZAVAK: NEUTRON DIFFRAKCIÓ, NEUTRON SZÓRÁS, ANYAGVIZSGÁLATI MÓDSZEREK, MÁRVÁNY, KERÁMIA, BRONZ

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The discovery of ancient artefacts and artwork usually raises a variety of questions such as the correct determination of their historical and cultural timeframe, the place and method of production, the choice of treatments and conditions for restoration and preservation.

In the field of archaeometry, new perspectives are opened up by the use of neutron techniques such as Neutron Diffraction, SANS and USANS (Small/Ultra-Small Angle Neutron Scattering) or recently neutron tomography. Neutron is a suitable probe for investigating the interior of cultural heritage objects. Neutrons penetrate through materials with different degree of attenuation, depending on the atomic component of the sample. This property makes neutrons ideal for non-destructive testing. Information on the artefacts are relative to composition, presence of alteration, crusts, inclusions, structure of the bulk, typology of the location of materials extraction, manufacturing techniques, refactoriness, porosity and firing temperature. Provenance and state of conservation of stone objects are of key importance, to address questions of attribution and restoration of archaeological objects.

Some of the questions archaeologists ask to archaeometrists deal with various aspects such as, the case of ceramic studies: to support the macroscopical (i.e. archaeological) fabric classification and grouping, to characterise the raw materials (clays and temper), to document production technology (shaping and firing techniques), to investigate provenance of the raw materials.

It is very important to link the aims and quality of the performed archaeometrical work to the concept of 'Classical Archaeology'. In general, this discipline was very much concerned with the more representative aspects of archaeology (sculpture, inscriptions and architecture) and paid insufficient attention to the potentiality of hand-craft studies, despite the pottery investigations has proved to address a wide range of questions, beyond providing the archetypical chronology. Artefact studies are a prerequisite to perform an all-round classical archaeology and in principle archaeometry forms part of that package. In fact, archaeometry is extremely useful in some cases, but we should always be aware of the fact that these techniques are adding a new layer of information or a new perspective.
Figure 1. A picture of Villa Adriana - Tivoli (Rome)

The answers are mostly very technical, however, and need interpretation. The outcome of analyses is not an end result, it is a good step on the way, but we should always be aware that research started with archaeology asking the question to archaeometry, and that in the end, conclusions need to be based on archaeological, rather than purely archaeometrical criteria.

A recent preliminary application of neutron techniques in the cultural heritage field is a neutron diffraction study of marbles from Villa Adriana in Tivoli (Rome) (Filabozzi, A et al. 2006a, 2006b) (see Figs. 1, 2). Villa Adriana is an exceptional complex of classical buildings designed and erected in the 2nd century AD by the Roman Emperor Hadrian, and is recorded by the UNESCO in the World Heritage List. Analytical investigations on monumental complexes of the Roman Empire complement the studies carried out on architectural and building engineering in order to achieve unitary views on this historical period. In this context, neutron diffraction technique has been used for the characterisation of ancient Roman marble fragments from the 'Edificio con Tre Esedre' palace, located in Villa Adriana (n.22 in Fig. 2).

Figure 2. Map of Villa Adriana.
This technique produced complementary information compared to conventional study of XRD, etc. The marble decoration was built with the 'opus sectile' technique, typical of Hadrian's time, with wall decorations belonging to the 2nd century AD. A reconstruction of the marble wall decoration of the Palace 'Edificio con Tre Esedre' is shown in Fig. 3. Fragments come from the first comb rubbing and have been chosen as representative sampling of the discovered marbles and are shown in Fig. 4.

Marble is one of the most common stones used for monuments, statues and other objects of archaeological interest and is typically composed of either calcite or dolomite or a combination of the two. This study aimed at distinguishing different marble types on the basis of the mineral phase compositions and the crystallographic textures. Information obtained can be used to identify the origin of the different marbles.

The Diffraction experiments on marbles were performed on the time-of-flight (TOF) diffractometers GEM and ROTAX at ISIS (UK). The use of a multi-detector TOF neutron diffractometer allows performing quantitative analysis on stationary samples in a matter of minutes. This technique has many advantages: a stationary experimental set-up to collect a complete diffraction pattern can be used, objects of variable shapes or sizes can be illuminated without prior preparation, and preferred orientation or texture effects are easily recognised.

Multi-phase and multi-bank analysis of the diffraction patterns were carried out with standard crystallographic public-domain software (GSAS [http://www.ccp14.ac.uk] for quantitative phase analysis and MAUD [http://www.ing.unitn.it/~maud] for texture analysis). An example of experimental data for marble of n.11, together with the final GSAS Rietveld refinement is shown on Fig. 5. Phase analysis performed on diffraction measurements allow to classify the marble fragments into 4 groups: one contains only calcite, the second contains calcite and illite/muscovite, a third calcite, quartz and traces of illite/muscovite and the forth contains "non-marble" samples.
The texture information is displayed in the pole figures. Pole figures are the 'maps' of the crystal grains orientations, determined in marbles by tectonic processes as creation, deformation, geological processes and so on, and can be used as fingerprints to identify them. On Fig. 6 a sketch of a pole figure reconstruction from the diffraction measurements (left) together with texture reconstruction along (006) for marble fragments n. 9, 19, 22,... (right), are shown. Pole figures indicate more or less pronounced deviations from a random crystallite orientation distribution (n.17).

At present, further studies on Roman pottery and Italian bronzes using diffraction and tomography techniques are in progress.

Roman pottery selected for the investigations (Fig. 7.) are part of a large recent collection of findings in Southern Italy (1988, Brindisi, Puglia). It provided a large number of material coming from different regions within the central Mediterranean (Greece, in particular) and eastern regions (Syria, Palestine, Asia Minor), dated 2\textsuperscript{nd} -3\textsuperscript{rd} century A.D.

The studies of Dean Arnold and David Peacock propose an innovative approach to the archaeological interpretation of ceramics which significantly extends our understanding of the social, cultural and environmental processes of ceramic production. They advocate a more integrated approach of pottery production mechanisms, taking not only ethnographical evidence into account, but also applying the full potential of archaeometrical analyses.

For instance for ceramics, that generally contain different quantities and kinds of impurities, like Hf, Ga, As, Ba, Sb, etc., a quantitative determination of such elements can provide information about the geographical area of provenance (Buxeda I Garrigós et al. 2002). In fact the results could provide information about the site where the clays were extracted and on the manufacturing method.

The bronze samples are findings form Southern Italy (Sicily) dated 6\textsuperscript{th} -5\textsuperscript{th} century BC. They are small statues, of unknown bronze casting technique.
Figure 7.
Cnidian Relief decorated vases (oinochoe) in Lecce University, Laboratorio di archeologia classica, coming from Brindisi (Santa Chiara)

These objects, in the ancient past or in the 17th century, have probably undergone alterations or insertion of non-original parts.

Neutron diffraction and tomography technique could answer non-destructively these questions, through the quantitative determination of the constituent phases, their proportions in the sample, and on crystallographic texture of the most abundant phases.

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