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Texture and structure studies on marbles from Villa Adriana via neutron diffraction technique

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The primary objective of this work is the characterisation of ancient roman marble fragments through neutron diffraction, a non-destructive experimental method. The neutron diffractometer ROTAX, operating at the pulsed neutron source ISIS, in the UK has been used to determine composition down to a 0.5 wt% level and to obtain information on preferred orientations of grains in the marble tiles.

Q3 *Keywords*: Ancient Roman marbles; Neutron diffraction; Textures; Muscovite–illite

1. Introduction

In this paper a neutron diffraction characterisation of ancient roman marble fragments is presented. The samples originate from Villa Adriana (Tivoli, Rome, Italy), an exceptional complex of classical buildings designed and erected in the 2nd century AD by the Roman Emperor Hadrian and which is inscribed by UNESCO in the World Heritage List. The analytical investigation on the monumental complexes of the Roman Empire Age complements the studies carried out on architectural and building engineering in order to achieve unitary views on this historical period and the present investigation on microscopical structure of the marble fragments. This work is carried out within the RiVA (Rivelare Villa Adriana) initiative, a joint project involving the University of Rome Tor Vergata and the Ministry of Cultural Heritage, aiming, among other things, at a thorough chemical–physical analysis of the large variety of Villa Adriana artistic artefacts, in a multidisciplinary context. Q4 The marbles investigated in the present work come from the red and green areas of the. The neutron diffraction experimental technique is a very effective tool for the non-destructive and bulk analysis of archaeological objects at the microscopic scale, where no drilling, coring, cutting, scraping are required and thus ideal for their characterisation [1–3].

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The neutron probe presents many advantages such as high penetration in the material (several cm), a good and fine sampling (i.e. characterisation may regard the whole artefact), and it requires a simple and stationary experimental set-up.

Marble is one of the most common stones used for monuments, statues and other objects of archaeological interest in the Villa. In this context the provenance and the state of conservation of stone objects are of key importance. The quantitative phase analysis resulting from the diffraction study provides a picture where the set of marbles investigated shows a composition typically of either calcite or dolomite or a combination of the two. A quantitative phase analysis has been carried out in order to identify mineral components down to a 0.5 wt% level. Texture analysis with neutron diffraction has also been used to obtain information on preferred orientations of grains in the marble tiles and fragments and will serve as a fingerprint characteristic for a particular type of marble. The type and the strength of texture, determined by neutron diffraction, is used to identify the origin of the different marbles. Indeed it has been proposed that part of the artefacts might have originated from quarries in the Mediterranean area of known textures.

2. Experimental

Measurements have been performed on the ROTAX neutron diffractometer (ISIS, UK) The set-up of the instrument is shown in figure 1. Time-of-flight (TOF) measurements are used to determine neutron energies. The instrument is designed for high-resolution (thus allowing sharp diffraction peaks) measurements on thick samples and to operate with mostly stationary experimental set-ups (i.e. no sample or detector movements). The latter guarantees that diffraction patterns can be collected at any detector angle so that orientation or texture effects are easily recognised.

In this experiment marble samples (dimensions: 2–6 cm × 1–3 cm × about 5 mm thick) were mounted onto a goniometer installed inside an evacuated tank. The diffraction patterns were recorded for 21 different marble samples, with a measuring time of about 1 h each (figure 2). A set of texture measurements for eight of the marble samples have also been performed. The marble sample was rotated in 144 different orientations in order to map it. The diffraction pattern in backscattering was used to reconstruct the pole figures (figure 4).

The results of the GSAS Rietveld refinements [5,6] carried out on the diffraction data indicate that the samples investigated can be classified into four distinct categories: (A) only calcite; (B) calcite + illite/muscovite; (C) calcite + quartz + traces of illite/muscovite; (D) non-marble samples with no calcite or dolomite. Results of this analysis are reported in table I. For each identified phase a fraction of the mineral can be attributed, apart from the phase indicated as muscovite–illite, for which a structure model is not available. For this mineral we can say that for marbles of type C there are "traces" of phase muscovite–illite, while for marble type B it is present in significant quantities.

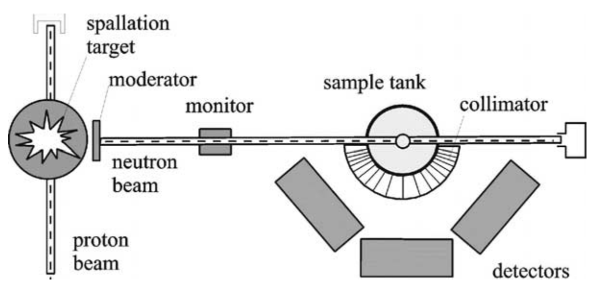

Figure 1. Experimental set-up of the TOF ROTAX diffractometer at ISIS.

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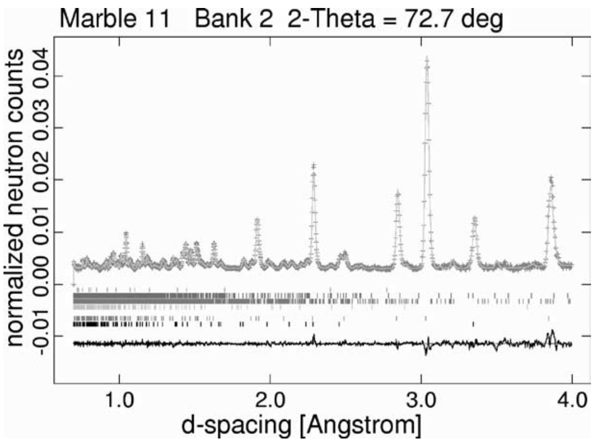

Figure 2. Diffraction pattern measured for sample no. 18, plotted as a function of *d*-spacing. It is pure calcite, as far as neutron diffraction data can tell.

Table 1. Identified phases of marble samples used in this experiment.

Type	*Sample nos.*	*% Calcite*	*% Quartz*	*% Dolomite*	*% Muscovite–illite*	*% Plagioclase*
A	2, 18, 22, 25, 29, 30	99.997	0.003			
B	1, 3, 5, 9	78.2–89.9	0.2–2.5	0.3–0.8	9.1–20.8 = significant	0–2.4
C	11, 16, 21	82.8–85.2	14.7–16.2	0.1–1.9	Traces	

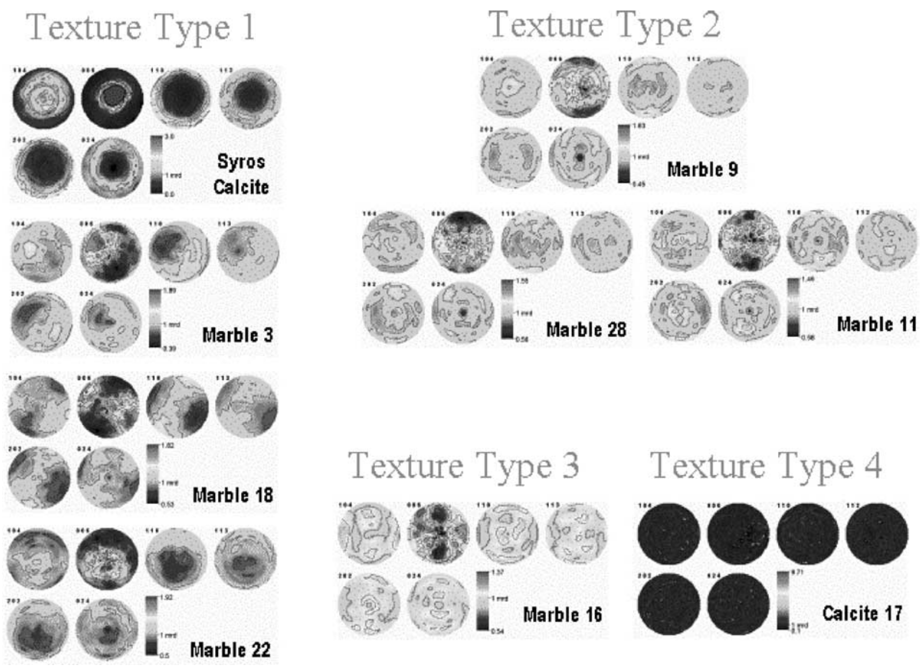

Figure 3. ROTAX diffraction patterns plotted as pole-figures: no texture is present in samples of type 4, while in the others marbles have experienced a *c*-axis compression.

Results of the texture measurements [7] are shown in figure 3. These show ROTAX diffraction patterns displayed as the "pole-figures", that is a map of the orientation distribution of crystallites in a polycrystalline material. Texture is a signature of the history of an object (creation, deformation, geological processes. . .) and can be used as a fingerprint to identify them [4] non-destructively: type and strength of texture are characterising features. When the grains are randomly oriented, the material is "texture-free". Samples of type 1 present pronounced density maxima in the (006) pole figure, and a girdle of pole density in the (110) pole figure, indicating a highly axisymmetric texture, as a result of a compression in the direction of the crystallograph *c*-axis. The texture strength is in this case quite high, indicating a strong deviation from a random distribution. The same symmetry, but a lot weaker, is present in samples of type 2 and 3. For type 4 texture is not present, indicating an almost random distribution of grain orientation.

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