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A Study of Various Types of Natural Marble Towards Their Use in Cultural Applications

Nikolaos Andreas Kazakis, Athena Research and Innovation Center in Information Communication and Knowledge Technologies, Greece

George I. Dallas, Athena Research and Innovation Center in Information Communication and Knowledge Technologies, Greece

Nestor C. Tsirliganis, Athena Research and Innovation Center in Information Communication and Knowledge Technologies, Greece

ABSTRACT

Marble has been used as decorative or building material since ancient times. Various archaeological marble materials of cultural heritage (e.g., statues) are frequently encountered in excavation sites amongst other finds. The scope of the present work is to investigate whether natural marble could be used for dosimetric purposes in cultural applications, such as dating or authenticity test of ancient marble objects, through an integrated experimental approach. For this purpose, the chemical composition, structure/morphology, and luminescence properties of 11 different types of natural marble of different origin (from various geographical regions) were examined by means of micro-x-ray fluorescence spectroscopy, stereoscopic microscopy, thermoluminescence (TL), and optically stimulated luminescence (OSL), respectively. Results indicate that though TL and OSL could be used for the identification of the marble type (origin), the use of natural marble for dosimetric purposes using these methods requires the a-priori knowledge of its type along with some of the main luminescence properties.

KEYWORDS

Chemical Analysis, Dosimetry, Marble, Optically Stimulated Luminescence, Origin, Thermoluminescence

INTRODUCTION

Marble has been used as decorative or building material since ancient times. Various archaeological marble materials of cultural heritage (e.g., statues) are frequently encountered in excavation sites amongst other finds. Thus, marble could possibly be used for dosimetric purposes in cultural applications, such as dating or authenticity test of ancient marble objects.

The suitability of luminescence techniques has been investigated for this purpose, as extensively employed to date archaeological ceramic materials and geological samples. More specifically, Thermoluminescence (TL) and Optically Stimulated Luminescence (OSL) can be used for dating marble materials (e.g., Liritzis et al., 1997; Liritzis and Galloway, 1998; Theocaris et al., 1997; Polikreti et al., 2003; Liritzis, 2010) when certain requirements/assumptions are met. More specifically, dating of marble objects (i.e., when the marble was lastly exposed to sunlight) can be accomplished providing that their (external) surface has been exposed to daylight for a long period of time before being exploited (Liritzis, 2010). Otherwise, incomplete bleaching will leave a residual dose in the material

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providing a highly erroneous age result if it is not subtracted from the archaeological dose (Liritzis et al., 1997). In addition, TL has also been applied in marble authenticity testing (e.g., Theocaris et al., 1997; Polikreti et al., 2002; 2003), but more effort is needed towards this direction (Polikreti, 2007).

Polikreti (2007) also notes that for any type of stone, there is no physicochemical method to ensure reliable answers to authenticity and/or dating problems, especially in the case of marble. This is attributed to the fact that marble is a geological material, thus dating techniques would give its geological age. The history of the object is recorded only at the external layer, which however is affected by numerous factors (e.g., marble type, grain size, porosity, environmental conditions, etc.). Thus, each marble surface constitutes a separate case, with its own characteristics (Polikreti, 2007).

Moreover, marble is a highly inhomogeneous metamorphic rock, which can be found in various forms and colors exhibiting different properties, strongly depending on its origin. To this respect, Khamis and Arafah (2017) made a preliminary study presenting the variations of the TL-glow-curve shape for different types of natural marble.

Based on the above, the scope of the present work is to study eleven (11) different types of natural marble of different origin (from various regions) through an integrated approach. All marble types are investigated by means of micro-X ray fluorescence spectroscopy (mXRF), stereoscopic microscopy, TL and OSL. The above measurements shed light on the similarities and variations in their chemical composition, structure/morphology and luminescence properties respectively based on their origin. Results are then assessed to further evaluate the suitability and/or the drawbacks of using natural marble as a reliable tool for dosimetric purposes in cultural applications (e.g., dating of ancient buildings or marble objects) and/or other applications (e.g., accidental dosimetry).

MATERIALS, INSTRUMENTS AND METHODS

Eleven different types of natural marble of different origin were studied in the present work, which are presented in Table 1. Every type of marble has different characteristics, such as color, porosity, inclusions, etc.

Sample Preparation

The stereoscopic examination and the mXRF measurements took place on a small piece of each marble type without any pretreatment to gather a full understanding of the structure and chemical composition of their main bodies.

On the other hand, the marble samples required a special treatment before the luminescence measurements (OSL and TL). Small pieces of each marble sample were gently crushed using an agate mortar and then sieved to get different size fractions. The grains of 75–150 μm diameter were then heated at 60 ^{o}C for about 3 hrs to remove any water content. The dried grains were then placed (~3 mg) in stainless steel cups to prepare the aliquots for the OSL and TL measurements. The entire procedure took place under light conditions.

Stereoscopic Examination

Stereoscopic study of the marble samples was achieved by means of a high-performance stereoscopic microscope (Leica MZ-APO) with a maximum magnification of 80x. The entire imaging system was apochromatically corrected. A digital micro-camera (Leica DC 200) was also attached to the stereoscopic microscope and connected to a computer allowing the on-line viewing, processing and storage of the acquired images through an appropriate software. The micro-camera had an optical resolution of 1920×1536 (3.0 Mpixels), while illumination and color correction of the images were also controlled through the software.