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Towards the Discrimination between Natural and Synthetic Pigments: The Case of Ultramarine

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Ultramarine has been for centuries one of the most highly prized pigments of all traditional artists'materials, due to its durability, excellent color, and its intrinsic value. For the production of the blue pigment Ultramarine, the rare semiprecious stone Lapis Lazuli is used, which has been mined since ancient times in mines northeast of Afghanistan, making it difficult to transport to the Mediterranean region. This accounts for the pigment being named ultramarine blue, i.e. a blue pigment coming from beyond the sea. The production of a synthetic version by Guimet in 1828, which was obtained from the calcination of a mixture of metakaolin, sulfur, sodium carbonate, and a reducing agent, followed by an oxidation step, introduced an important change in artists' habits, in that a less expensive pigment was available for their palettes [1]. The verification of the natural or synthetic origin of the pigment is particularly important in a project, e.g. in authentication cases.

In this research work, ten samples of natural and synthetic Ultramarine pigments were used to investigate the possibility of their discrimination using characterization methods. In the first stage of the research, all samples were studied by Fourier transform infrared (FTIR) spectroscopy. The distinction between natural and synthetic ultramarine using FTIR -as proposed in previous studies- is difficult, as the FTIR bands of interest coincide with the peaks of atmospheric CO_2 ($2400 - 2300 cm^{-1}$) [2,3]. For this reason, X-ray diffractometry (XRD), X-ray photoelectron spectroscopy (XPS), scanning electron microscopy (SEM-EDS), and UV-Vis spectrophotometry were used for all samples to support the study [4,5]. The combination of elemental composition, morphological analysis, phase identification, chemical state, color calculation, and optical band gap estimation is studied, to propose an analytical protocol for the discrimination between samples of artificial and natural origin.

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Primary authors: POURLIAKA, Anastasia (School of Physics, Faculty of Sciences, Aristotle University of Thessaloniki); MALLETZIDOU, Lamprini (School of Physics, Faculty of Sciences, Aristotle University of Thessaloniki); KARFARIDIS, Dimitrios (School of Physics, Faculty of Sciences, Aristotle University of Thessaloniki); ZORBA, Triantafyllia (School of Physics, Faculty of Sciences, Aristotle University of Thessaloniki); PARASKEVOPOULOS, Konstantinos M. (School of Physics, Faculty of Sciences, Aristotle University of Thessaloniki); VOURLIAS, George (School of Physics, Faculty of Sciences, Aristotle University of Thessaloniki)

Presenter: POURLIAKA, Anastasia (School of Physics, Faculty of Sciences, Aristotle University of Thessaloniki)

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