

Carbon speciation of paints with mixed protein-oil binders: an inelastic x-ray scattering approach

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The transition from egg-based tempera to oil painting in quattrocento was a turning point in art history and influenced the creation of Renaissance masterpieces by renowned artists of the time. Sandro Botticelli, Domenico Ghirlandaio, and Leonardo da Vinci continued to use egg in parallel or in combination with oil and prepared paints of great complexity. Protein-oil mixtures, used as paint binders, can result in diverse microstructures based on the sample preparation procedure [1,2,3]. Today, we are still missing critical information on the chemical composition and microstructure of such mixed media paints. Here, we present x-ray Raman scattering (XRS) spectroscopy as an element-sensitive bulk probe for disentangling complex chemical speciation of organic mixed-media paints.

In the XRS process, an incident photon is inelastically scattered by a core electron, and part of its energy is transferred to excite the inner-shell electron into an empty state, providing x-ray absorption near edge structure (XANES) data comparable to conventional x-ray absorption techniques [4]. In our recent studies, XRS has been explored as a powerful spectroscopic tool for the chemical characterization of a natural-occurring organic collection of plant exudates [5] and as a bulk non-invasive 3D spectral imaging probe for the identification and discrimination of carbon-based compounds in organic paleontological specimens [6]. In this work, we identify and compare the carbon bonding of paints prepared in diverse ways (i.e., oil, egg tempera, tempera grassa (egg with oil), oil paints with protein-coated pigments). We also discuss the main limitations of the approach (i.e., spatial resolution, radiation-induced effects), and we suggest complementary techniques and mitigation strategies to overcome them.

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[2] Ranquet, Ophélie, et al. "A holistic view on the role of egg yolk in Old Masters' oil paints." *Nature Communications* 14.1 (2023): 1534.

[3] Ranquet, Ophélie, et al. "Tempera and Tempera Grassa: From Wet Paints to Solid Films." *ACS Applied Polymer Materials* 5.7 (2023): 4664-4677.

[4] Georgiou, Rafaella, et al. "X-ray Raman Scattering: A Hard X-ray Probe of Complex Organic Systems." *Chemical Reviews* 122.15 (2022): 12977-13005.

[5] Georgiou, Rafaella, et al. "Disentangling the chemistry of Australian plant exudates from a unique historical collection." *Proceedings of the National Academy of Sciences* 119.22 (2022): e2116021119.

[6] Georgiou, Rafaella, et al. "Carbon speciation in organic fossils using 2D to 3D x-ray Raman multispectral imaging." *Science Advances* 5.8 (2019): eaaw5019.

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