Probing Complex Organic Heritage Materials with X-ray Raman Scattering

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The analytical means to assess the organic composition of complex samples are limited and precious for heritage studies. The work unfolds the potentials of x-ray Raman scattering (XRS) as a non-invasive probe for the study of paleontological, archaeological and art materials. We optimize the XRS methodology required for the study of complex samples, including organic paleontological specimens and a collection of plant exudates of heritage value, and explore its ability to access the different levels of information sought. XRS is employed as a bulk, non-invasive, 3D imaging probe to study ancient organic samples to determine organic signatures and trace their evolution through time. Taking advantage of the direct tomography technique based on chemical bond contrast, we introduce 3D x-ray Raman imaging (XRI) in the field of natural heritage studies. The successive collection of XRS carbon K-edge tomographic images of an exceptionally preserved specimen, a 53 million-year old insect entrapped in amber, provides a means to assess the spatial distribution of biological remnants. The results point to the importance of characterization of paleontological specimens through imaging at the scale of the object, to understand the specimens' biochemistry, molecular evolution, and interactions between the organism and the depositional setting. Three-dimensional chemical imaging appears essential for understanding chemically altered heterogeneous heritage materials that have undergone diagenesis and aging processes over time. We demonstrate that high energy resolution XRS spectroscopy provides a new perception in the analysis of organic cultural heritage materials. We examine a historical collection of Australian plant exudates which assembles natural products used by Aboriginal peoples for thousands of years. Detailed analysis of the spectral signatures of core electron transitions in the carbon and oxygen atoms, allows us to identify chemical bonds and inter-compare plant secretions belonging to distinct chemical classes (terpenoids, polysaccharides, phenolic and aromatic compounds). We exemplify via statistical analysis that characterization and discrimination of the complex organic compounds is attained using XRS in a non-invasive manner. Additional statistical approaches have been developed to process the three-dimensional XRS imaging data and to decompose the obtained XRS spectra. We show that scanning transmission x-ray microscopy (STXM), in the soft x-ray range, complements XRS approach when its spatial limit is reached. We have analyzed organic dark-coloured pigments documented in De Mayern's manuscript by STXM. The nanoscale images provide new insights on the carbon chemistry of these pigments and demonstrate their chemical complexity that can only be addressed at the nanometric spatial resolution. In conclusion, the results exemplify the value of XRS as an in-situ non-invasive spectroscopic and imaging probe of light elements in heritage materials. Through this work we provide a new viewpoint for exploitation of XRS data in a wide range of organic, heterogeneous, and chemically complex systems, and discuss its remaining challenges and future perspectives.

THÈSE

Les membres du jury sont :

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Vous êtes cordialement invités au pot qui suivra

Dans la limite de 25 personnes et dans le respect des gestes barrières

Formalités d'entrée : accès libre limité à 25 personnes maximum ou en visioconférence

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