The Scientific Legacy of Prof. Georges Calas: Applications of Synchrotron Radiation Methods to Earth Materials, Environmental Geochemistry, Mineral Resources, and Nanomaterials

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The first applications of very intense x-rays from synchrotron radiation (SR) sources to earth science problems were made in the late 1970's and early 1980's. Prof. Georges Calas was one of the pioneers of these applications, particularly the use of SR-based x-ray absorption fine structure (XAFS) spectroscopy to determine the speciation (oxidation state and local structural environments) of first-row transition elements in silicate glasses (*e.g.*, Calas and Petiau, *Bull. Mineral.*, **1983**, *106*, 33). Since then, there have been many applications of XAFS and other SR-based methods, such as x-ray fluorescence imaging, to determine the speciation of elements at concentrations ≥50 ppm in crystalline and amorphous solids and in complex mixtures of minerals and natural organic matter in coals and oil shales as well as in aqueous fluids and silicate melts at high temperature. The use of XAFS to determine sorption modes of heavy metal and actinide species at mineral-aqueous solution interfaces (*e.g.*, Hayes *et al.*, *Science* **1987**, *238*,783) helped spawn the field of molecular environmental science (*e.g.*, Morin *et al.*, *Am. Mineral.* **1999**, *84*, 420). SR-based x-ray microscopy (*e.g.*, STXM and x-ray ptychography) enable even higher spatial resolution imaging (≥15 nm) and XAFS in the soft x-ray energy range (100 to 2400 eV) on natural samples containing microbial organisms (*e.g.*, Benzerara *et al.*, *GCA* **2008**, *72*, 3949). X-ray free electron lasers now allow ultra-fast XAFS studies that can follow pathways of chemical reactions, including the first detection of transition-state complexes (Oström *et al.*, *Science* **2015**, *347*, 978). We will review selected applications of SR to molecular-scale problems involving earth materials, complex environmental samples, sorption processes, nanominerals and their transformations in S-rich environments, and dissolution and precipitation of minerals in complex geological samples.