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Imaging Chemistry (and Physics) in Space and Time: New Opportunities by Dynamic Neutron and X-ray micro-Imaging

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Reactive transport phenomena in porous media are of fundamental relevance not only in hydrology or environmental sciences, but also in a broad range of other scientific disciplines as well as in numerous engineering processes. For most chemically active porous materials or composite systems, their physical structure and chemical reactivity is directly reflected in the spatial distribution of chemical properties. Key chemical properties include chemical composition (distribution of chemical elements), chemical speciation (atomic coordination of a specific chemical element), or chemical states (e.g. redox state). Chemical information of this kind provides key knowledge about chemical reactivity as well as structural properties of heterogeneous materials. Consequently, current demands on imaging extend beyond traditional structural (physical) imaging. The ability to visualize the distribution of chemical properties at the (sub)micrometer scale as well as chemical dynamics in materials and reactions is of fundamental interest and importance. Thus, the need for „chemical microscopes"is growing rapidly.
Over the most recent years, micro-analytical facilities based on neutron beams as well as synchrotron x-ray beams advanced to indispensable instruments in the context of micro-analytical, non-destructive imaging. Both types of beams are nowadays used for multi-dimensional structural micro-analysis (physical imaging) as well as chemical and crystallographic micro-imaging. Advantageously, based on their characteristic and generally non-destructive interaction with matter, neutrons and x-rays provide complementary analytical contrast mechanism.
X-ray microprobe facilities posses several intrinsic advantages concerning chemical imaging. Most important, the element-specific absorption resonances accessible within the x-ray energy range provide an element-specific chemical sensitivity. Moreover, the fine structure of an absorption edge reveals information on the chemical speciation of the absorber. Quite commonly, different oxidation states are readily identified based on their characteristic spectroscopy signatures of the absorption edges. Furthermore, any specific chemical environment around the absorber (chemical speciation) will lead to specific bound-bound transitions at the low-energy side of the edge, while the coordination geometry will result in characteristic scattering phenomena above the Fermi level. These speciation-dependent features represent chemical contrast which can be used to record up to three-dimensional chemical images documenting the spatial variation of oxidation states, specific mineral phases, or different molecular species, for example.
Complementary to x-rays, neutron radiation exhibits a unique penetration power and a particular sensitivity for lowest Z elements. Neutron radiography and tomography is the most suitable non-destructive tool for dense objects when a certain size of the object is reached. Compared to the more common X-ray approaches, even heavy elements can be penetrated and distinguished from other materials. Moreover, due to the large cross section of low Z elements, small amounts of aqueous liquids or organic materials can be detected with a high contrast even within a dense body. Neutron imaging corresponds indeed to a highly specialized method allowing the observation of hidden structures and features in bulk objects.

In this presentation, recent progress and achievements in the field of 2D/3D chemical imaging and speciation analysis using various neutrons and synchrotron radiation x-ray microprobe techniques will be demonstrated. The reactive transport of Cs in natural Opalinus Clay Rock material will serve as an example of scientific application. Clearly, Opalinus Clay Rock represents a chemically and physically heterogeneous medium. The complementary use of different microprobe techniques allows characterizing the physical structure and chemical nature of the porous medium. Moreover, the effect of the observed heterogeneities on the evolution of reactive transport pattern can be visualized. Most advantageous, the employed neutron and x-ray techniques can be considered as being of non-invasive nature (in most cases, at least). This important feature allows the non-disturbing recording of multiple, subsequent images as a function of evolution –making up a "reactive transport movie". In the presented case, we used such time-resolved chemical imaging to conduct dynamic, in-situ investigations elucidating the diffusion of water and an inert tracer, as well as the mobility of reactive aqueous species. Notably, the evolution of Cs contaminant plumes in various micro-structured natural porous media differing in physical and chemical complexity could be monitored.
Finally, through a model based interpretation of experimental results we grasp the mechanisms of transport phenomena and chemical interactions in porous media. In this context, the rapid development of analytical imaging techniques goes along with novel data analysis and modeling approaches. Available time resolved

3D chemical imaging and structure analysis opens opportunity for advanced 3D transport modeling taking into account compositional heterogeneity of the samples at a (sub)-micrometer scale consistently with the experimental observations.

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