Fingerprinting reaction pathways of lightweight compounds by X-ray Raman scattering Spectroscopy: Application to hydrogen storage materials

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In situ and in operando studies of chemical reactions by X-ray absorption spectroscopy allows for precise characterization of reaction products and reaction intermediates via fingerprinting by reference compounds [1]. As the spectral shape of the absorption edge is highly sensitive to the chemical environment of the absorbing atom, e.g. its oxidation state, local coordination, spin state and type of bonding, multicomponent fits of known references can be utilized to determine changes in the phase composition during reaction. However, this method is limited to the study of high Z elements whenever the sample environment (chemical reactor, high pressure cell, electrochemical cell) or the reaction conditions (pressurized gas atmosphere, solid-liquid interfaces) cause strong absorption of X-rays. In those cases X-ray Raman scattering offers a powerful approach to study absorption edges of low and intermediate Z elements bulk sensitively [2] and is particularly valuable for the analysis of amorphous reaction products and liquid phases. Thanks to the recent development of large solid-angle spectrometers, in situ / in operando X-ray Raman scattering experiments became feasible nowadays [3,4] which opens an exciting field for new applications in e.g. catalysis, reaction kinetics, battery research, and fuel cell technology. The capabilities and limitations of the method will be discussed and exemplified by the study of decomposition pathways in the potential lightweight hydrogen storage materials $Mg(BH_4)_2$ and $Ca(BH_4)_2$, during thermally induced hydrogen release [5-7].

References

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