Operando REELS of hydrogen selective membrane systems

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In a future scenario with hydrogen as an energy carrier, the separation and purification of the energetically costly hydrogen will become even more important than today.[1] Here, we make use of an experimental model system (consisting of a V thin film coated Pd membrane) to study material combinations for hydrogen selective membranes, in which the surface properties of the system can be modified intentionally.[2] The *in situ* preparation allows for observation of ultra-clean hydrogenation and the quantification of the permeation kinetics and the surface elemental characterization by XPS/AES. We determine the (sub-) surface hydrogen concentration by *operando* reflecting electron energy loss spectroscopy (REELS) as a function of pressure and temperature, from which the surface hydrogen pressure-composition isotherms are derived (Figure 1). The energy dependence of REELS reveals that VH_x exhibits a hydrogen concentration gradient. We demonstrate the existence of a rate-limiting step by direct observation of the dependence of the permeation on the hydrogen content in the Pd/V composite membrane. Modelling of the permeation behaviour yields activation energies in good agreement with other hydrogen absorbing thin film systems [3] for the overall process, the dissociation at the Pd surface, the diffusion in the membrane, and the dissociation at the V surface.

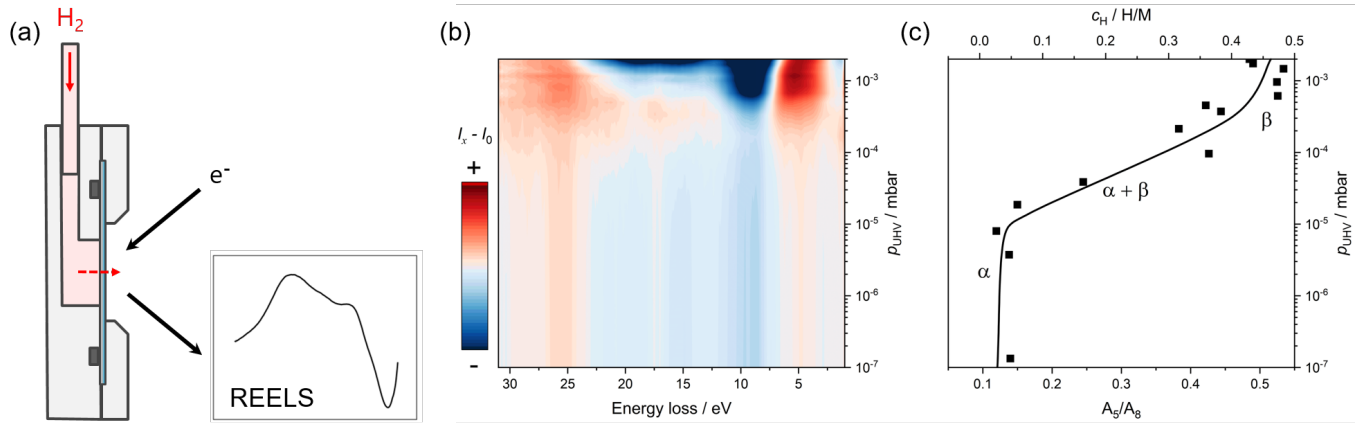

Figure 1: (a) Schematic representation of the membrane setup for electron spectroscopy. (b) REELS difference spectra of VH_x (x = 0 – 0.3) recorded at 100 °C under increasing hydrogen pressure from which a surface hydrogen pressure-composition isotherm (c) can be derived.

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Acknowledgements
Financial Support from the Swiss National Science Foundation (Grant No. 172662) and the UZH-URPP *LightChEC* is greatly acknowledged.