Electroanalytical methods for monitoring solvent induced changes in diffusion and reaction kinetics - The case study of hydrogenases in redox hydrogels.

Redox hydrogels (highly solvated polymers films) were developed as matrices for the integration of redox enzymes in bioelectrochemical devices toward technological application. High loading in protein is possible via physical entrapment within the hydrogel and finely tuned redox relays allow for fast electron transfer at low overpotential (see for example [1]). The solvated environment preserves the activity of the immobilized enzyme. In addition the hydrogel properties can be tuned to provide a protection mechanism for the biocatalysts [2] or serve as a confined matrix. In particular hydrogenase, are highly active catalysts for H_2 evolution or oxidation, but suffer from deactivation induced by oxygen and can be stabilized in redox hydrogels films.

Solvation of the redox hydrogels defines electron transfer, mass transport and catalytic activity of the films and can be tuned by variation of the solvent and co-solutes. For example, changes in the nature and concentration of the counter-ions provide a simple way for changing the solvation state of the hydrogel. Electroanalytical methods provide insights into the changes in electron transfer, mass transport and catalysis and hence allows for fine tuning of the solvent properties. Hydrogenase embedded in redox hydrogels will serve as a case study to identify rate limiting steps and the solvent effect via electroanalytical methods.

References

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[2] N. Plumeré, O. Rüdiger, A. Alsheikh Oughli, R. Williams, J. Vivekananthan, S. Pöller, W. Schuhmann, W. Lubitz, *Nature Chemistry*, **2014**, *6*, 822–827.