Nanoelectrodes in Picoleter Droplets: Towards Single Molecules Chemistry

The aim of this project is to understand the behaviour of small ensembles of molecules – ultimately down to single molecules. We are focused on the oxygen reduction reaction, which is important in biology as well as in a number of technical systems. Special attention is paid to the deactivation of enzyme molecules and short-lived reactive oxygen species. These species decompose very quickly or react rapidly with other components of solution.

We will use electrochemical techniques as they allow, in contrast to optical techniques, the observation without high power illumination. We use the dispensing of tiny droplets of electrolyte solution onto nanoscopic electrode arrangements. Both are even 100 times smaller than human hair thickness. Such systems provide the sensitivity required for activity analysis of single enzyme molecules over time or to achieve a total electrochemical conversion of droplet components within short time. The microscopic liquid phase of the droplet prevents short-lived species against diffusional escape or spreading and allows the build-up of measurable concentrations with a low amount of converted substances. We will address two aspects: the formation and follow-up reactions of reactive oxygen species, and the catalytic activity of a small number of enzyme molecules. The nanoscopic dimension of the new type of electrochemical cell will enable the detection of short-lived species before they decompose or diffuse to the solution bulk. We would like to follow activity of individual enzyme molecules over time in order to understand their deactivation (gradual vs. stepwise).

New analytical methodologies proposed within this project will allow their utilization in studies of other systems comprising single entities and short-lived species. A direct observation of reactive oxygen would open new possibilities to study specific reaction of these short lived entities with solution components, biological and technical systems. Understanding the deactivation mechanisms would open new doors for the rational design of environments or reaction conditions under which the activity of enzymes can be better preserved to make them more useful for technical application.