Adsorption simulations of biomolecules on SAM coated surfaces

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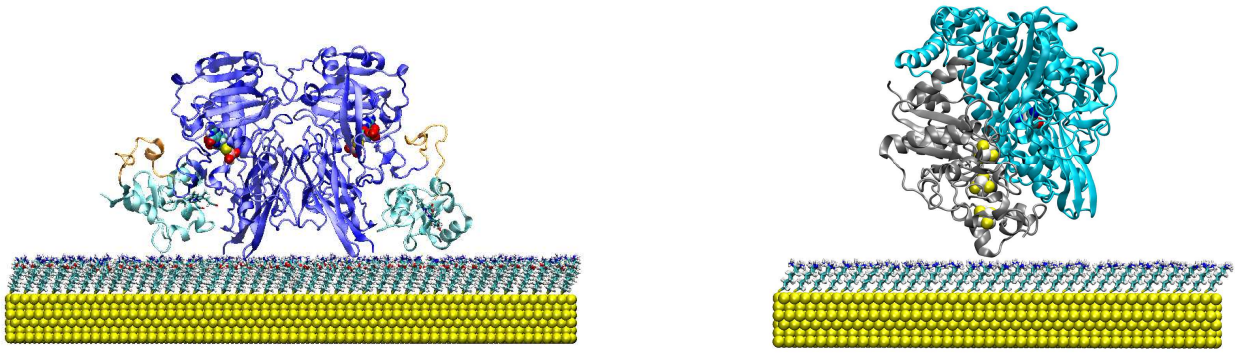

The adsorption of biomolecules onto surfaces is an important issue in many fields. Here, we apply classical molecular dynamics simulations complemented by spectroscopy to investigate the immobilization of the sulfite oxidase and [NiFe] hydrogenases, which are of interest for the development of biosensors and biofuel cells, respectively.

For the sulfite oxidase catalyzing the oxidation of sulfite to sulfate, it was demonstrated that the adsorption process is strongly ionic strength dependent. While under low ionic strength the flexibility of the enzyme is strongly restricted upon immobilization, it stays more mobile under high ionic strength conditions [1]. This observation is of special interest, because predicted domain motion events have a strong effect on the electron transfer [2].

[NiFe] hydrogenases catalyze the reversible cleavage of hydrogen into protons and electrons. They are grouped into oxygen sensitive (standard) and tolerant hydrogenases.

In a first study, the electrostatically driven adsorption of standard hydrogenases was investigated by changing the protonation level of the self-assembled monolayer (SAM) coating the gold surface [3]. We observed that higher charge densities on the SAM led to a stronger immobilization, but affected protein stability beyond a certain value.

Furthermore, the immobilization of the oxygen tolerant membrane bound [NiFe] hydrogenase (MBH) was probed. This adsorption process was more challenging, because the MBH contains an additional membrane anchor and a much weaker dipole moment. Therefore, the influence of the anchor and different surfaces was studied [4]. The work showed that both, the anchor and the surface constitution, had strong effects on the immobilization.

[1] T. Utesch et al., *Langmuir*, **2012**, *28*, 5761-5769.
[2] A. Pacheco et al., *J. Biol. Inorg. Chem.*, **1999**, *4*, 390-401.
[3] T. Utesch et al., *Langmuir*, **2013**, 29. 673-682
[4] T. Utesch et al., *(in preparation)*