Artificial transfer hydrogenase encapsulated in silica nanoparticles

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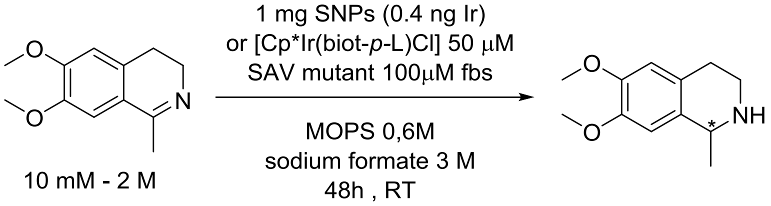
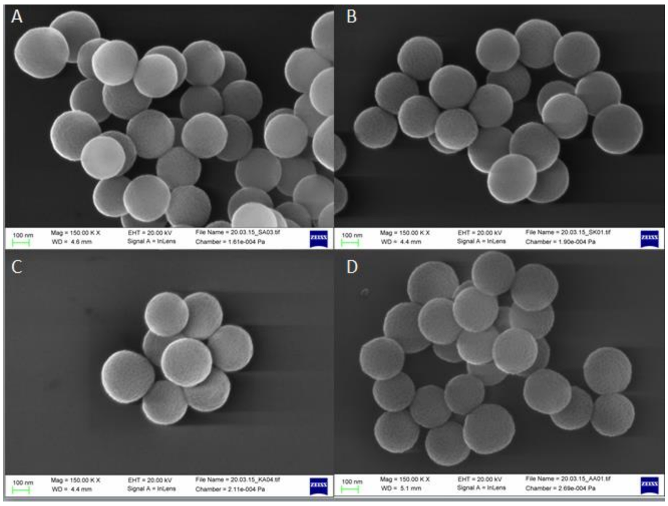
An artificial metalloenzyme (ArM) is a hybrid catalyst created by incorporation of an organometallic cofactor within a host protein scaffold.[1,2] This system, based on the biotin-streptavidin technology, combines organometallic and enzymatic catalysis,[3,4] and can therefore catalyze multiple chemical processes. With the aim of immobilizing Artificial Transfer Hydrogenase (ATHase) on silica nanoparticles (SNPs) and performing NAD$^+$ regeneration *in situ*, we selected the enantioselective transfer hydrogenation of cyclic imines as a model reaction.



The lyophilized streptavidin isoforms were dissolved in water and pre-incubated with the biotinylated iridium cofactor. The solutions were then lyophilized to obtain the functional artificial metalloenzymes, which were embedded within a protective organosilica layer [5] in collaboration with the group of Prof. Shahgaldian yielding active SNPs.



Upon immobilization and protection of streptavidin mutants on the SNPs, we were able to obtain over 47 000 TON for the substrate reduction, with the possibility to recycle the active catalyst. This protected system is also able to retain its activity in the presence of various cellular debris without any treating agent. The concentration of the active Cp*Ir catalyst was determined by means of ICP-MS measurements.

References

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