Artificial Metalloenzymes: (Strept)avidin as Host for Enantioselective Hydrogenation by Achiral Biotinylated Rhodium−Diphosphine Complexes

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We report on the generation of artificial metalloenzymes based on the noncovalent incorporation of biotinylated rhodium−diphosphine complexes in (strept)avidin as host proteins. A chemogenetic optimization procedure allows one to optimize the enantioselectivity for the reduction of acetamidoacrylic acid (up to 96% ee (R) in streptavidin S112G and up to 80% ee (S) in WT avidin). The association constant between a prototypical cationic biotinylated rhodium−diphosphine catalyst precursor and the host proteins was determined at neutral pH: log Ka = 7.7 for avidin (pI = 10.4) and log Ka = 7.1 for streptavidin (pI = 6.4). It is shown that the optimal operating conditions for the enantioselective reduction are 5 bar at 30 řC with a 1% catalyst loading.
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