Artificial metalloenzymes based on biotin-avidin technology for the enantioselective reduction of ketones by transfer hydrogenation

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Most physiological and biotechnological processes rely on molecular recognition between chiral (handed) molecules. Manmade homogeneous catalysts and enzymes offer complementary means for producing enantiopure (single-handed) compounds. As the subtle details that govern chiral discrimination are difficult to predict, improving the performance of such catalysts often relies on trial-and-error procedures. Homogeneous catalysts are optimized by chemical modification of the chiral environment around the metal center. Enzymes can be improved by modification of gene encoding the protein. Incorporation of a biotinylated organometallic catalyst into a host protein (avidin or streptavidin) affords versatile artificial metalloenzymes for the reduction of ketones by transfer hydrogenation. The boric acidůformate mixture was identified as a hydrogen source compatible with these artificial metalloenzymes. A combined chemo-genetic procedure allows us to optimize the activity and selectivity of these hybrid catalysts: up to 94% (R) enantiomeric excess for the reduction of p-methylacetophenone. These artificial metalloenzymes display features reminiscent of both homogeneous catalysts and enzymes.
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