638d A Localized Enantioselective Catalytic Site on Short DNA Sequences and Their Amphiphiles

Details

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About

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DNA-based artificial metalloenzymes (ArMs) combine the chiral environment of the DNA scaffolds and the catalytic activity of transition metals and have demonstrated high enantioselectivity for a wide range of reactions. However, despite progress in correlating the catalytic performance of DNA-ArMs with their DNA sequence, pinpointing which of the many possible chiral microenvironments created by the interaction between the DNA and the bound metal complex is responsible for enantioselective catalysis, remains elusive. Here, in order to address this question, we have designed a series of double-stranded DNA (dsDNA) sequences that bind to the copper (II) complex of 4,4'-dimethyl-2,2'-bipyridine (dmbipy-Cu). Our results demonstrate that a DNA-ArM based on dmbipy-Cu and a dsDNA as short as 8 base pairs with only two contiguous central G•C pairs (G for guanine and C for cytosine), is sufficient for the catalysis of a highly enantioselective Diels-Alder reaction in water. Molecular dynamics simulations indentified dissimilar sequence-dependent binding patterns for the dmbipy-Cu to the short dsDNA. In addition, when the short dsDNA was conjugated to hydrophobic tails, the resulting DNA-ArM exhibited enantioselective properties in a methanol-water mixture superior to that of non-amphiphilic dsDNA, as well as dsDNA-amphiphiles with more complex G•C-rich sequences.

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