

Effect of the ordered hydration layer for the enzymatic reactions on DNA scaffold

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The catalytic enhancement of DNA scaffolded enzymes have been attributed to the reduced adsorption,¹ lower local pH² or ordered hydration layer on the DNA scaffold surface,³ however, the exact working mechanisms remain unclear. In this study, xylose reductase (XR) and xylitol dehydrogenase (XDH), with the optimal activity at pH 6 or 8 equally displayed significant catalytic enhancements on the DNA scaffold. By using a pH sensor, a lower local pH shift of 0.8 was observed near the DNA scaffold. The postulated local pH change near the DNA scaffold surface unlikely plays a general role in enhancing the activity of scaffolded enzymes. Instead, the ordered hydration layer attracted by the negatively charged DNA scaffold surface is a plausible candidate for the general factor for the catalytic enhancement.⁴

Here, we hypothesized that the ordered hydration layer enhanced the activity of DNA scaffolded enzymes by increasing the local concentration of hydrophilic substrates. This hypothesis was tested by the hydrophilic or hydrophobic substrates of XR, and the of salts (NaCl or CsCl) in a high concentration that disrupted the ordered hydration layer. To further tested this hypothesis, the enzyme reactions of carbonic anhydrase (CA) in free and DNA scaffolded form were investigated with different hydrophobic substrates. This study gives the new insight into the origins of the catalytic enhancement of enzymes assembled on the DNA scaffold.

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