Spatially Organized Enzymes Drive Cofactor-Coupled Cascade Reactions

We report the construction of an artificial enzyme cascade based on the xylose metabolic pathway. Two enzymes, xylose reductase and xylitol dehydrogenase, were assembled at specific locations on DNA origami by using DNA-binding protein adaptors with systematic variations in the interenzyme distances and defined numbers of enzyme molecules. The reaction system, which localized the two enzymes in close proximity to facilitate transport of reaction intermediates, resulted in significantly higher yields of the conversion of xylose into xylulose through the intermediate xylitol with recycling of the cofactor NADH. Analysis of the initial reaction rate, regenerated amount of NADH, and simulation of the intermediates’ diffusion indicated that the intermediates diffused to the second enzyme by Brownian motion. The efficiency of the cascade reaction with the bimolecular transport of xylitol and NAD(+) likely depends more on the interenzyme distance than that of the cascade reaction with unimolecular transport between two enzymes.

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