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Constructing spatially separated multienzyme system through bioadhesion-assisted bio-inspired mineralization for efficient carbon dioxide conversion

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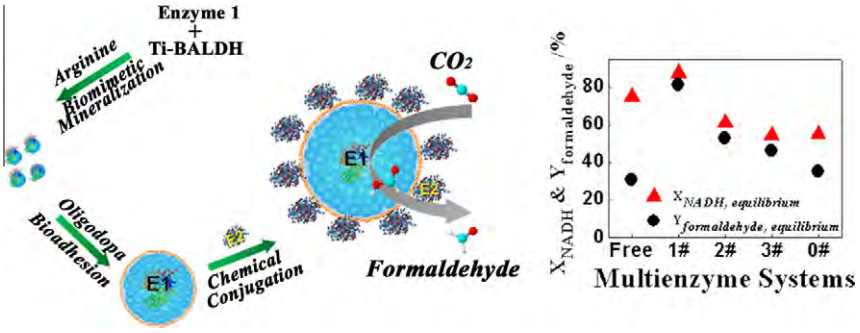
HIGHLIGHTS

- A bioadhesion-assisted bio-inspired mineralization approach is developed.
- A spatially separated multienzyme system is constructed.
- The multienzyme system displays size-related high activity and stability.

GRAPHICAL ABSTRACT



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ABSTRACT

A facile and green bioadhesion-assisted bio-inspired mineralization (*BABM*) approach is proposed to construct spatially separated multienzyme system for conversion of carbon dioxide to formaldehyde. Specifically, formate dehydrogenase is entrapped accompanying the formation of titania nanoparticles (NPs) through bio-inspired titanification. After *in situ* surface functionalization of NPs with oligodopa, formaldehyde dehydrogenase is immobilized on the surface of NPs through amine-catechol adduct reaction. Compared to co-immobilized and free multienzyme system, the spatially separated multienzyme system exhibits significantly enhanced formaldehyde yield, selectivity and initial specific activity. The influence of particle size on the enzyme activity reveals that the formaldehyde yield (80.9%, 52.9%, 46.4%), selectivity (92.7%, 86.6%, 85.1%) and initial specific activity (1.87, 1.31, 0.29 U mg^{-1}) all decreased as the NPs particle size increased from 75, 175 to 375 nm. After storing for 20 days at 4 °C, this multienzyme system retains as high as 70% of its initial activity.

1. Introduction

Considering the energy and environmental urgency, the development of greener and more efficient catalytic conversion processes has emerged as one of the major goals of modern chemical engineering and bioresource technology (Noyori, 2009). Among the existing catalytic conversion process, multienzyme catalysis (*i.e.* the combination of two or more enzymatic transformations in concurrent one-pot processes), is particularly appealing since enzymes are intrinsically "green" biological catalysts for a broad variety of catalytic reactions, which could provide industrially important products with an excellent chemo-, regio-, and stereo-selectivity (Santacoloma et al., 2011; Mamma et al., 2008). Moreover, the multienzyme catalysis process has some intrinsic advantages: (Ricca et al., 2011) (1) the demand of time, cost and chemicals for product recovery could be reduced and (2) the concentration of harmful or unstable compounds could be decreased

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