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Short Communication

Cascade enzymatic reactions for efficient carbon sequestration

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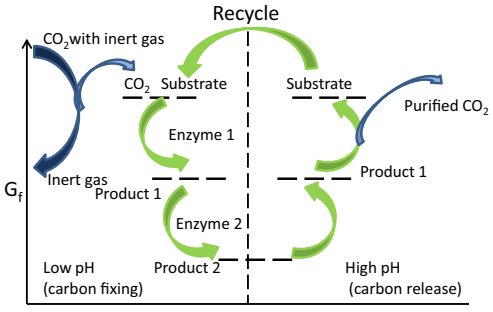
HIGHLIGHTS

- Reversible cascade enzyme reactions promise highly efficient carbon capture.
- Manipulation of thermodynamic driving forces for carbon conversion via pH control.
- Coupled cascade enzymic reactions greatly enhanced carbon capture capacity.
- Biocatalytic capture of 0.5 mol CO_2 with each mole of substrate applied.

GRAPHICAL ABSTRACT



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ABSTRACT

Thermochemical processes developed for carbon capture and storage (CCS) offer high carbon capture capacities, but are generally hampered by low energy efficiency. Reversible cascade enzyme reactions are examined in this work for energy-efficient carbon sequestration. By integrating the reactions of two key enzymes of RTCA cycle, isocitrate dehydrogenase and aconitase, we demonstrate that intensified carbon capture can be realized through such cascade enzymatic reactions. Experiments show that enhanced thermodynamic driving force for carbon conversion can be attained via pH control under ambient conditions, and that the cascade reactions have the potential to capture 0.5 mol carbon at pH 6 for each mole of substrate applied. Overall it manifests that the carbon capture capacity of biocatalytic reactions, in addition to be energy efficient, can also be ultimately intensified to approach those realized with chemical absorbents such as MEA.

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1. Introduction

Carbon capture and storage (CCS) has been pursued as the most promising strategy in recent years to mitigate carbon emission from our still steadily-growing consumption of fossil fuels. Through a CCS process, CO_2 is captured and separated from its gaseous mixtures (such as flue gas from coal-fired power plants) into a pure gas, which can be subsequently liquefied and injected into underground storage sites (Boot-Handford et al., 2014). Alternatively, the purified CO_2 can also be converted into value-added products, or applied to various industrial processes (Cheng et al., 2011). It has been reported that CCS processes have reached a level of 2 billion ft^3 CO_2/yr in oil recovery industry by the end of 2010 (Leach et al., 2011). Most industrial CCS processes have been realized using chemical absorbents. Among other options, amines such as monoethanolamine (MEA), diethanolamine (DEA) and methyldiethanolamine (MDEA) were the most common agents for industrial processes (Rochelle, 2009). Such weak basic chemicals

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generally react with CO_2 at low temperatures (~50 °C), realizing the separation of CO_2 from its gaseous mixtures; the resulted products are required to be decomposed subsequently, thus regenerating the absorbents and releasing CO_2 as a pure gas. However, chemical regeneration requires elevated temperatures, mostly above 120 °C. Since the process involves large amount of water (~80% w/w in the solution), large portions of the energy are wasted during the heating–cooling cycles. To seek energy-efficient CCS strategies, growing interests have been shown in biological processes such as the growth of micro algae (Choi et al., 2012; Kumar et al., 2011). Our interests have been directed to *in vitro* biocatalytic carbon capture (Tong et al., 2011; Baskaya et al., 2010; Xia et al., 2014), which can be operated in a way similar to chemical carbon sequestration but promises high energy efficiency as the regeneration and reuse of substrates can be realized at ambient conditions; the reactions can also be readily intensified through well-developed reaction engineering means.

Generally speaking, biocatalytic reactions afford lower carbon capture capacity than chemical reaction routes, if compared based on the amount of substrate/absorbent applied for each reaction cycle. Due to the basic nature of the chemical absorbents, chemical carbon capture reactions can be driven to almost complete conversion, while most biocatalytic reactions (typically carboxylation reactions) reach much lower degree of conversion as limited by reaction equilibrium. Considering that carbon fixation has been achieved through cascade reactions in biology, we assume herein that such reactions conducted simultaneously in a homogeneous solution can further the conversion of substrate, thus improving the overall carbon capture capacity. Indeed, cascade reaction have been reported successful for conversion of CO_2 to methanol, both enzymatically (El-Zahab et al., 2008; Baskaya et al., 2010) and chemically (Huff and Sanford, 2011).

Isocitrate dehydrogenase (ICDH) and aconitase were two key enzymes constituting the reductive tricarboxylic acid cycle (RTCA) for carbon fixation. ICDH catalyzes the conversion of CO_2 and ketoglutarate to isocitrate, which is further converted to citrate by aconitase. The reversibility of these individual reactions has been demonstrated through previously reported studies (Beh et al., 1993; Xia et al., 2014; Colman, 1975). The current work examines the potentials, especially the thermodynamic equilibrium limitations and feasibilities for such cascade reactions for carbon sequestration when applied in a one-pot manner.

2. Methods

2.1. Materials

α-Ketoglutaric acid, NADPH, D-isocitrate, sodium bicarbonate, 2-(N-morpholino ethanesulfonic acid (Mes), N,N-bis(2-hydroxyethyl)glycine (Bicine), monopotassium phosphate, dipotassium phosphate, aconitase from porcine heart (A5384) were purchased from Sigma Chemical Co. (St. Louis, MO, USA). The plasmid pET28A and *Escherichia coli* strain BL-21(DE3) were purchased from Novagen (Bilerical, MA, USA). All other chemical agents were purchased and used with the highest purity available.

2.2. Enzyme expression, purification and characterization

The gene of ICDH from *Chlorobium limicola* was synthesized and cloned into pET28A plasmid (Novagen, USA), and the resultant recombinant plasmid was expressed in *E. coli* BL-21(DE3) (Novagen, USA). ICDH was purified by Ni-affinity chromatography. Purified enzyme was kept in pH 7 phosphate buffer and the concentration of enzyme was measured with Bradford assay.

Aconitase (purchased from Sigma) was activated before being used. Typically, 15 mg of the enzyme was dissolved in 2.0 ml 0.1 M Tris buffer (pH 7.4). The solution was then mixed with 50 μl of 1 mM ferrous ammonium sulfate (0.39 mg/ml in water) and 0.1 ml of 0.05 M L-cysteine (7.90 mg/ml in water, adjusted to pH 7.4 with NaOH). The mixture was then incubated for 1 h at 0 °C before being used.

2.3. Enzyme specific activity assay

For ICDH, one unit of activity was defined as 1 μmol of NADPH produced (decarboxylation) or consumed (carboxylation) per min at room temperature. For decarboxylation, the activity assay reaction mixture contained 1 mM isocitrate, 2 mM NADP+, 40 mM $MgCl_2$, and 20 μg/ml enzyme dissolved in 1 mL of buffer solution. Mes buffer was used for pH 6.0–7.0 solution, while Bicine buffers for pH 8.0–9.0. The reaction solution was monitored continuously with a UV–Vis spectrometer (50 Bio, Varian) for changes in the concentration of NADPH via absorbance at 340 nm. For carboxylation, the same reaction conditions were applied with substrates and additives changed to 10 mM ketoglutarate, 200 μM NADPH, 50 mM $KHCO_3$ and 40 mM $MgCl_2$.

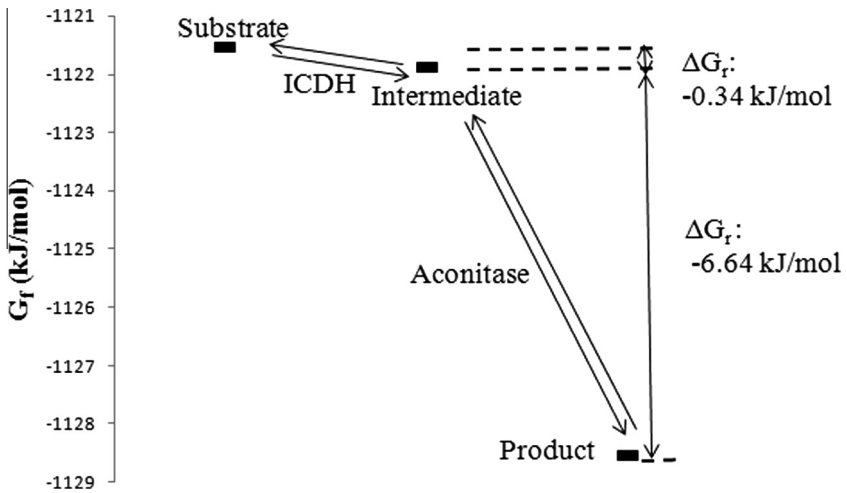

Fig. 1. Thermodynamic driving forces (Gibbs free energy) of reactions catalyzed by ICDH and aonitase (data were calculated using Debye–Huckel model for pH 6; data of G_f shown in the figure are for substrate and product mixtures).

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