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Biotransformation of formaldehyde (C1) into erythrulose (C4) by ThDP-dependent Carbon-Carbon Ligases without by-product formation

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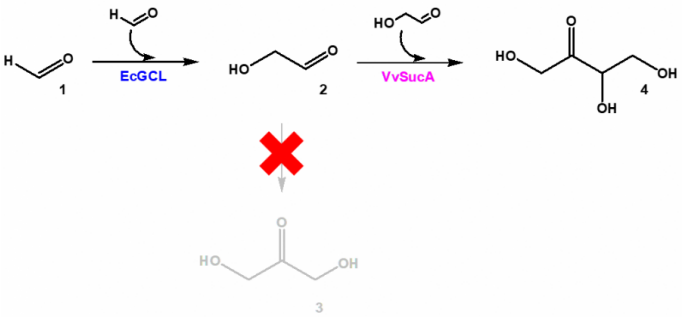
PURPOSE OF THE ABSTRACT

Erythrulose (C4) is used as an active ingredient for tanning in the cosmetic industry [1]. Thereby, we examined an enzyme cascade reaction system to selectively produce C4 compounds (e.g., erythrulose) from C1 carbon source (e.g., formaldehyde) without by-product formation (e.g., dihydroxyacetone) (Fig. 1). The enzyme cascade consisted of two thiamine-diphosphate dependent carbon-carbon ligases. The thermostable glyoxylate carboligase from Escherichia coli K-12 (EcGCL), which is able to condense two molecules of formaldehyde into one molecule of glycolaldehyde [2,3], was selected as the first enzyme. The EcGCL was engineered to improve catalytic efficiency based on the crystal structure with glycolaldehyde.

The SucA, a decarboxylating E1 subunit of the α-ketoglutarate dehydrogenase complex of Vibrio vulnificus (VvSucA) was chosen as the second enzyme (i.e., C2 carboligase), because it catalyzed the condensation of two molecules of acetaldehyde into one molecule of acetoin [4]. The coupling of EcGCL mutant (i.e., EcGCL_R484M/N283Q/L478M/M488L /R284K) and VvSucA_K228L allowed to produce 0.9 g/L erythrulose from 3 g/L formaldehyde via glycolaldehyde (Fig. 2). By-products (e.g., dihydroxyacetone) were not observed. This study will contribute to valorization of C1 gas into industrially relevant multi-carbon products in an environment-friendly way.

FIGURES



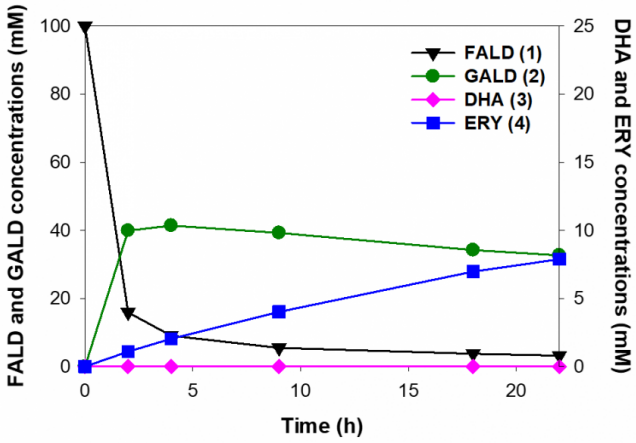


FIGURE 1

Biocatalytic pathway for producing erythrulose (3) from formaldehyde (1) via glycolaldehyde (2).

Erythrulose (4) can be made from formaldehyde (1) without dihydroxyacetone (3) by combination of EcGCL and VvSucA.

FIGURE 2

Biotransformation of formaldehyde (1) to erythrulose (4) via glycolaldehyde (2) by coupling EcGCL R484MN283QL478MM488LR284K and VvSucAK228L

KEYWORDS

Erythrulose | Formaldehyde | Carboligase | Enzyme cascade

BIBLIOGRAPHY

1. Samed, G., Vanessa, W., Andr, P., & Volker, S., Green Chem., 2021, 23, 6583-6590.
2. Jo, H. J., Kim, J. H., Kim, Y. N., Seo, P. W., Kim, C. Y., Kim, J. W., Yu, H. N., Cheon, H. J., Lee, E. Y., Kim, J. S., & Park, J. B., Green Chem., 2021, 24(1), 218-226.
3. Jo, H. J., Yu, H. N., Cheon, H. J., Kim, J. H., Kim, G. Y., Kim, B. S., Kim, J. S., & Park, J. B., ACS Sustainable Chem. Eng., 2023, 11(3), 1078-1086.
4. Seo, P. W., Jo. H. J., Hwang, I. Y., Jeong, H. Y., Kim, J. H., Kim, J. W., Lee, E. Y., Park., J. B., & Kim, J. S, Catal Sci Technol., 2020, 10, 79-85.