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TOPIC(s): Homogenous, heterogenous and biocatalysis

Evolved thermostable Transketolase for the valorization of vegetable oils

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PURPOSE OF THE ABSTRACT

The production of biosourced aliphatic aldehydes from vegetable oils, renewable raw material, offers many applications in the fields of cosmetics, detergents, polymeric materials and additives (Figure 1). Chemical processes by oxidative cleavage or in the presence of organocatalysts such as analogs of thiamine have been reported.[1,2] These processes require very high temperature, organic solvents and generate toxic by-products.

The goal of this study is to develop a novel ecofriendly enzymatic C-C bond cleavage process catalyzed by a thiamine diphosphate (ThDP) dependent enzyme, transketolase (TK) (Figure 2). This enzyme is commonly used for the stereoselective formation of a C-C bond in the presence of hydroxypyruvate as donor substrate, rendering the reaction irreversible, and an aldehyde as acceptor substrate allowing the valorization of ketose products.[3] Our project aims to exploit the reverse TK-catalyzed reaction for the C-C bond cleavage of a modified fatty acid 1 derived from methyl oleate as donor substrate in order to generate highly valuable aldehydes 2.

For this study, the thermostable TK from Geobacillus stearothermophilus (TKgst) [4] will be used with donor substrates of increasing complexity, from TK natural substrates such as fructose-6-phosphate to methyl oleate derivative 1. Since the wild-type enzyme preferentially accepts polyhydroxylated substrates, efficient TKgst variants already identified against hydrophobic compounds will be used.[5,6] These hydrophobic compounds might be difficult to solubilize in water media leading to the use of cosolvents or hydrotropes. Finally, different enzymatic cascade strategies will be investigated to shift the equilibrium of TK-catalyzed-reaction. The first results will be presented and discussed.

FIGURES

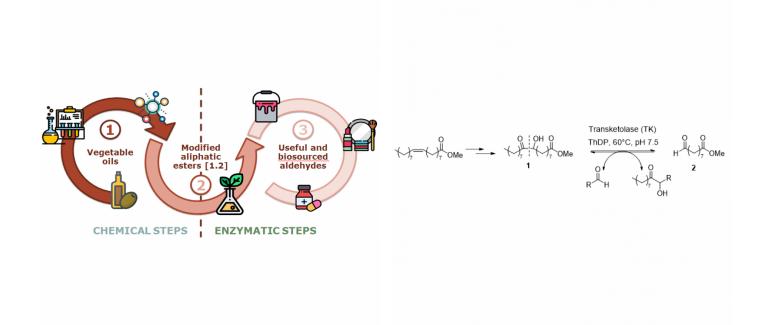


FIGURE 1

Project goal

FIGURE 2

Main route to fatty aldehydes 2 from a modified methyl oleate derivative 1

KEYWORDS

transketolase | aliphatic aldehyde | vegetable oils

BIBLIOGRAPHY

- [1] e. deruer, n. duguet and m. lemaire, chemsuschem, 2015, 8, 2481-2486.
- [2] n. d. vu, s. bah, e. deruer, n. duguet and m. lemaire, chem. eur. j., 24, 8141–8150.
- [3] y. kobori, d. c. myles and g. m. whitesides, j. org. chem., 1992, 57, 5899-5907.
- [4] j. abdoul-zabar, i. sorel, v. hélaine, f. charmantray, t. devamani, d. yi, v. de berardinis, d. louis, p. marlière, w.-d. fessner and l. hecquet, adv. synth. catal., 2013, 355, 116–128.
- [5] t. saravanan, s. junker, m. kickstein, m.-k. link, j. ranglack, s. witt, m. lorillière, l. hecquet, w.-d. fessner, angew. chem. int. ed. 2017, 56, 5358–5362
- [6] d. yi, t. saravanan, t. devamani, f. charmantray, l. hecquet and w.-d. fessner, chem. commun., 2015, 51, 480–483.