

N°715 / OC / PC

TOPIC(s) : Enzyme production, immobilization / Biocatalytic cascade reactions

SELECTIVE SYNTHESIS OF INDUTRY RELEVANT PRODUCTS BASED ON 5-HYDROXYMETHYLFURFURAL (HMF) CATALYZED BY SELF-SUFFICIENT IMMOBILIZED MULTI-ENZYME SYSTEMS

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PURPOSE OF THE ABSTRACT

Production of 2,5-dihydroxymethyl furan (BHMF) via the reduction of 5-hydroxymethyl furan (HMF) through an enzyme based biocatalytic process is a promising and more sustainable alternative to the manufacturing of building blocks for plastic production, which is normally based on fossil raw materials [1]. The replacement of traditional processes to produce plastics with the polymerization of BHMF obtained via enzymatic reaction is a clear path to a sustainable and environmentally friendly system without having to deny the use of plastics as a cheap and easily available material.

The biosynthesis of BHMF starting from bio-based HMF is challenging when using enzymes as biocatalysts due to the need of a high input of cofactor, which is depleted extremely fast. In this work, we have tackled this limitation via the co-immobilization of a redox cofactor (NADH) and two dehydrogenases capable of orthogonally reducing the HMF and recycling NADH in situ and inside the porous structure of the solid supports. This approach has allowed recycling both enzymes and cofactor for consecutive batch cycles.

YjgB Ec LND [2], an alcohol dehydrogenase from *Escherichia coli* is the enzyme in charge of the reduction of HMF into BHMF, while the thermostable glucose dehydrogenase developed by Figueroa [3] was chosen as the counterpart capable of delivering a cofactor recycling reaction. Both dehydrogenases were sequentially immobilized on a survey of supports to find the most optimal carrier and immobilization conditions that allowed reusing the co-immobilized enzymatic system over sequential HMF reduction/oxidation reactions. Purolite ECR8204F chemically modified with E-Co²⁺ and glyoxyl [4] groups turned out to be the best choice from such intense immobilization screening. The two enzymes achieved an immobilization yield of over 99% during the co-immobilization over E-Co²⁺ and glyoxyl carriers, and recovering 1.2 U/grsupport and 1.4 U/grsupport in the case of YjgB Ec LND, respectively for each carrier and 2.14 U/grsupport and 0.8 U/grsupport respectively in the case of the GDH.

This system is able to quantitatively produce BHMF by the reduction of HMF, regenerating the NADH in situ. The solid biocatalyst was reused up to fifteen times, in consecutive reaction courses of 1 h each, substituting the reaction media every cycle, and no decrease in conversion capabilities were detected along these cycles. The resulting bi-functional heterogeneous biocatalysts was further coated with cationic polymers capable of binding cofactor into their matrix, bringing the system one step closer to the true meaning of a self-sufficient catalyst, where exogeneous supply of cofactor is no longer needed and both enzymes and cofactors integrated into the solid carrier can be recycled simultaneously.

FIGURES

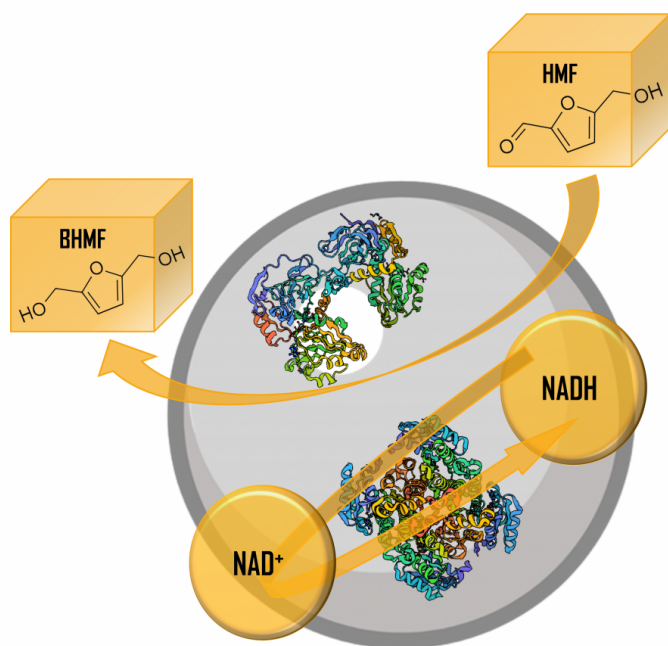


FIGURE 1

Scheme of the solid biocatalyst

Graphical representation of how the solid biocatalytic system that includes both alcohol and glucose dehydrogenases, is able to reduce HMF into BHMF at the same time as it is recycling the available cofactor (NADH).

FIGURE 2

KEYWORDS

Enzyme immobilization | Alcohol dehydrogenase | Glucose dehydrogenase | HMF reduction

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