

中空纤维膜固定化甲酸脱氢酶催化 CO₂ 合成甲酸

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摘要 以紫外光表面接枝改性的聚乙烯 (PE) 中空纤维膜为载体, 采用共价结合的方式固定化甲酸脱氢酶 (FDH), 考察了 CO₂ 通入方式、溶液 pH 值、缓冲液种类和还原型烟酰胺腺嘌呤二核苷酸 (NADH) 的浓度对酶催化 CO₂ 合成甲酸反应的影响。结果表明, 与加压法相比, CO₂ 鼓泡法更有利于甲酸的生成; 磷酸盐缓冲液优于 Tris-HCl 和盐酸三乙醇胺缓冲液; 体系 pH 值对反应的影响较大, 固定化 FDH 的最佳 pH 值仍为 6.0, 但 pH 耐受性增强; 随着辅酶 NADH 浓度的增加, 反应初速度加快, 收率下降; 游离酶和固定化酶的最大酶活分别为 0.246 和 0.138 mmol/(L·h); 固定化 FDH 在 4 °C 贮存两周后活性仅下降 4%, 而游离酶活性下降 50%。FDH 催化膜重复利用 10 次后, 活性没有明显降低。

关键词: 甲酸脱氢酶 中空纤维膜 固定化 酶催化 二氧化碳 还原

Abstract: Formate dehydrogenase (FDH) was covalently attached to surface-modified polyethylene (PE) hollow fiber membrane. The effects of inlet way of CO₂, pH value, the type of buffer solution, and the concentration of the reduced coenzyme nicotinamide adenine dinucleotide (NADH) on the synthesis of formic acid from CO₂ were investigated. The catalytic performance of PE-supported FDH and its reusability were studied. The results showed that the CO₂ bubbling method was superior to pressing method. Phosphate buffer was more favorable to the formation of formic acid than Tris-HCl and triethanolamine-HCl buffer. Immobilized FDH was less sensitive to pH than free FDH, and the optimum pH value was 6.0 for both systems. With the increase of NADH concentration, the initial reaction rate ascended while the yield tended to decrease. Enzyme activity reached 0.246 and 0.138 mmol/(L·h), respectively, for free and immobilized FDH when using 100 mmol/L of NADH. The stability was greatly improved after immobilization. The activity of PE-attached FDH only decreased by 4% while that of free enzyme dropped to 50% of initial activity after stored in phosphate buffer at 4 °C for two weeks. Furthermore, immobilized FDH exhibited outstanding reusability, which almost kept original activity after undergoing 10 cycles.

Keywords: formate dehydrogenase, hollow fiber membrane, immobilization, enzymatic catalysis, carbon dioxide, reduction

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