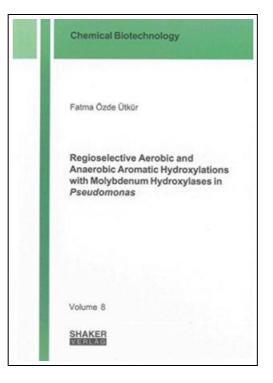
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Reviews

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REGIOSELECTIVE AEROBIC AND ANAEROBIC AROMATIC HYDROXYLATIONS WITH MOLYBDENUM HYDROXYLASES IN PSEUDOMONAS



Shaker Verlag Apr 2012, 2012. Buch. Book Condition: Neu. 21x14.8x cm. Neuware - Among many other interesting reactions, molybdenum (Mo) hydroxylases catalyze regioselective hydrocarbon oxyfunctionalizations, for which typically oxygenases are employed in synthetic applications. However, oxygenase-based processes are often limited by oxygen mass transfer, cofactor regeneration, and/or enzyme instability due to the formation of reactive oxygen species. As Mo-hydroxylases produce, rather than consume reducing equivalents during substrate hydroxylation and use water, rather than molecular oxygen as oxygen donor, these enzymes have a high potential for overcoming limitations encountered with oxygenases. The potential and feasibility of these enzymes for preparative applications was investigated in the frame of this thesis with quinoline 2oxidoreductase (Qor) and quinaldine 4-oxidase (Qox) serving as model enzymes. Up-to-date, several Mo-hydroxylases have been described, but rarely applied on industrial scale. For specific quinaldine hydroxylation to 4-hydroxyquinaldine, different Qox-based biocatalysts, reaction conditions, and key process parameters have been evaluated. The use of 1-dodecanol as carrier solvent and Qox-containing P. putida KT2440 as biocatalyst enabled high productivities (~0.4 g ltot -1 h-1) in a 0.5-L bioreactor setup without active aeration. Further evaluation of the key process parameters showed that inhibition by 1-dodecanol and the product was the most critical factor affecting process performance. As a proof of concept, the Qox-based process was coupled to downstream processing, including supercritical carbon dioxide treatment for breaking the stable emulsion followed by liquid-liquid extraction and crystallization allowing the isolation of 138 mg product with high purity (>99.9%). Furthermore, completely anaerobic quinoline hydroxylation was achieved with nitrate as electron acceptor using Qor-containing P. putida 86 expressing the nitrate reductase genes of P. aeruginosa. The achieved rate (7 U gCDW -1) shows the remarkable potential of Mohydroxylase-containing whole cells for O2-independent C-H oxyfunctionalizations. In conclusion, the development of an efficient integrated process and the first...

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