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Heterogeneous catalytic oxidation of phenol by in situ generated hydrogen peroxide applying novel catalytic membrane reactors

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Abstract
This work presents a novel method for oxidation of organic matter in water solutions based on catalytic membrane reactors. The oxidant, hydrogen peroxide, is generated directly in the bulk of the liquid investigated. Commercial symmetric alumina hollow fibers have been used as a starting material thereafter introducing the active phases. It has been proven that two different catalysts are necessary in order to complete the overall reaction, as well as to generate hydrogen peroxide and a heterogeneous Fenton process. Palladium has been used for the hydrogen peroxide generation and a second active phase, transitional metal oxides or homogeneous Fe^{2+}, has been used for the hydroxyl radical generation. An additional method for specific Pd loading to the reaction zone based on sputtering technique has been developed. All prepared catalytic membrane reactors (CMR) are capable of generating hydrogen peroxide in amounts comparable to CMRs reported in the literature. The catalytic membrane reactors prepared by Pd impregnation show very high activity and stability in phenol oxidation reaching 40% of the generated H_2O_2 usage in the oxidation reaction. Despite the very high activity of the catalytic membrane reactors obtained by Pd sputtering in H_2O_2 production they suffer very fast deactivation. Specific reactivation including a calcination step has been found to be appropriate for the recovery of their activity. Additional experiments give new insights for better understanding of Pd deactivation especially when the metal particles are of nanometer sizes.

Keywords
Hydrogen peroxide generation; Catalytic membrane reactor; Sputtered palladium nanoparticles; Phenol oxidation; Palladium nanoparticle deactivation

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