

Improving the sustainability and effectiveness of oxidative processes for water and wastewater purification

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# Summary

The growth of the population has occurred alongside the increase of industrial, agricultural and, correspondingly, waste production. It is no longer possible to evade the evidence of a tight nexus among human activities and the influence on the environment humans are part of. Indeed, the intensification of industrial activities turns into an unrelenting development of new classes of chemicals that are potentially harmful. If not properly managed, these substances may reach water resources through aqueous effluents, thus representing a risk for both the environment and human health. Moreover, the increasing demand of drinkable water worldwide follows the population growth. Access to safe water is not available everywhere and often untreated water sources are biologically contaminated. With this respect, oxidation processes are a promising tool against both chemical and biological contaminations of water effluents. This thesis investigation aims to increase the sustainability and the effectiveness of the classic Fenton oxidation process designing new and improved processes. A systematic approach is also presented to promote improved approaches for wastewater disinfection.

The first section of the investigation focuses on new strategies to thwart the diffusion of emerging chemical contaminants in water effluents. The identification of a clean, safe, versatile, efficient, cheap, and easy-to-handle approach is of crucial importance in water purification. The classic Fenton process is selected as a starting point for new developments in this direction. The use of effective iron ligands is the general strategy proposed to overcome some of the limitations of the Fenton process, namely, its inadequate selectivity, need to work under acidic pH conditions, undesirable by-products formation, sludge production, and high consumption of harmful compounds. A chitosan-iron system is proposed and efficiently applied as a polymeric organo-metallic catalyst in the degradation of chemical contaminants in water at circumneutral pH. A new and easy-to-use analytical method is discussed to discriminate among a metal-based and a free-radical oxidation mechanism, giving rise, respectively, to potentially selective and unselective processes. Selectivity is intended as both the ability to discriminate among different contaminants and the ability to favour oxidation of certain chemical positions in a contaminant. This work strongly suggests that an iron-ligand complex promotes a metal-based mechanism at circumneutral pH. Moreover, metabisulfite is proposed as an efficient and safer substitute of hydrogen peroxide as reactant in metal-based catalysed oxidation processes.

The second section of the investigation focuses on the impact of oxidative and non-oxidative biocides in water disinfection. The general objective of this section is to evaluate the real risk of the tested disinfectants to generate disinfection by-products, hence to define a strategy to limit disinfection by-products generation and promote a safer management of disinfectants. In this dissertation, disinfection by-products are generally intended as halogenated by-products. Study of the disinfection mechanism is the approach undertaken not only to understand the process, but to identify safer disinfection routes. Potential oxidants and non-oxidant disinfectants are investigated and applied in the disinfection of a real wastewater. Three safe biocides are identified, namely, thiazolinones, hydrogen peroxide, and metabisulfite. They are evaluated in terms cost-benefit analysis, efficiency toward biological contaminants, and environmental impacts. Moreover, an investigation of the mechanism of disinfection by-products generation in the use of peracetic acid is discussed. The study shows that peracetic acid, an increasingly adopted oxidant for wastewater disinfection, can act as organic substrate in the generation of bromoform if bromide is present in the aqueous matrix.

Overall, the results of this dissertation aim at promoting more sustainable processes for the removal of chemical and biological contamination from water. The study follows a general approach whereby investigation of the reaction mechanisms is performed to gain insight of the oxidation phenomena. This insight is in turn the basis of the exploration of systems with direct implications for engineering applications.