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Review

Electrochemical advanced oxidation processes: A review on their application to synthetic and real wastewaters

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ABSTRACT

Over the last decades, research efforts have been made at developing more effective technologies for the remediation of waters containing persistent organic pollutants. Among the various technologies, the so-called electrochemical advanced oxidation processes (EAOPs) have caused increasing interest. These technologies are based on the electrochemical generation of strong oxidants such as hydroxyl radicals (•OH). Here, we present an exhaustive review on the treatment of various synthetic and real wastewaters by five key EAOPs, i.e., anodic oxidation (AO), anodic oxidation with electrogenerated H_2O_2 (AO-H_2O_2), electro-Fenton (EF), photoelectro-Fenton (PEF) and solar photoelectro-Fenton (SPEF), alone and in combination with other methods like biological treatment, electrocoagulation, coagulation and membrane filtration processes. Fundamentals of each EAOP are also given.

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Abbreviations: AC, activated carbon; ACF, activated carbon fiber; AO, anodic oxidation; AOP, advanced oxidation process; BDD, boron-doped diamond; BOD_5, 5-day biochemical oxygen demand; CF, carbon felt; CNT, carbon nanotube; COD, chemical oxygen demand; CPC, compound parabolic collector; DOC, dissolved organic carbon; DSA, dimensionally stable anode; EAOP, electrochemical advanced oxidation process; EC, electrocoagulation; EF, electro-Fenton; GDE, gas-diffusion electrode; LMCA, low-molecular-weight carboxylic acids; MCE, mineralization current efficiency; MF, microfiltration; MWWTP, municipal wastewater treatment plant; NF, nanofiltration; PAN, polyacrylonitrile; PC, peroxi-coagulation; PEF, photoelectro-Fenton; PF, photo-Fenton; PTFE, polytetrafluoroethylene; RO, reverse osmosis; ROS, reactive oxygen species; RSM, response surface methodology; RVC, reticulated vitreous carbon; SPC, solar heterogeneous photocatalysis; SPEF, solar photoelectro-Fenton; SPF, solar photo-Fenton; SS, stainless steel; TOC, total organic carbon; TSS, total suspended solids; UF, ultrafiltration; UV, ultraviolet; UVA, ultraviolet A; UVB, ultraviolet B; UVC, ultraviolet C; UV–vis, ultraviolet-visible; VSS, volatile suspended solids; WWTP, wastewater treatment plant; $[TDI]_0$, initial total dissolved iron concentration; [TDI], total dissolved iron concentration; E, electrode potential (V); EC_V, energy consumption per unit volume ($kWh\,m^{-3}$); EC_{DOC}, energy consumption per unit DOC mass ($kWh\,(g\,DOC)^{-1}$); E_{cell}, average cell potential (V); E°, standard redox potential (V/SHE); I, current intensity (mA); j, current density ($mA\,cm^{-2}$); j_{cat}, cathodic current density ($mA\,cm^{-2}$); Q, specific charge ($Ah\,L^{-1}$); T, temperature (°C); V, volume (L); λ, wavelength (nm); •OH, hydroxyl radical; Cl•, chorine radical; HO_2•, hydroperoxyl radical; $SO_4^{\bullet -}$, sulfate radical.

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1. Introduction

One of the main current worldwide concerns is the growth of water pollution by organic compounds arising from many industrial, agricultural and urban human activities. The vast majority of these compounds are persistent organic pollutants, owing to their resistance to conventional treatments such as coagulation, biological oxidation, adsorption, ion exchange and chemical oxidation. As a result, they have been detected in rivers, lakes, oceans and even drinking waters all over the world. This constitutes a serious environmental health problem mainly due to their toxicity and potential hazardous health effects (carcinogenicity, mutagenicity and bactericidality) on living organisms, including human beings [1–4].

Over around the past three decades, research efforts have been made at developing more effective technologies to totally remove persistent organic pollutants from wastewaters. In this context, advanced oxidation processes (AOPs) acquired high relevance [5]. AOPs are based on the in situ production of highly reactive hydroxyl radicals ($^{\bullet}OH$) that non-selectively react with most organics, being able to degrade even highly recalcitrant compounds [6]. This radical is the second strongest oxidant known after fluorine, displaying a high standard redox potential of $E^{\circ}(^{\bullet}OH/H_2O) = 2.80$ V/SHE [7] and rate constants for reaction with several contaminants in the order of 10^6 to 10^{10} M^{-1} s^{-1} [8]. Moreover, $^{\bullet}OH$ have a short lifetime, estimated as only a few nanoseconds in water [9], and so they can be self-eliminated from the treatment system. The most common AOPs are H_2O_2 with UVC radiation (H_2O_2/UVC), ozone and ozone based processes (O_3, O_3/UVC, O_3/H_2O_2 and O_3/H_2O_2/UVC), titanium dioxide based processes (TiO_2/UV and TiO_2/H_2O_2/UV) and Fenton's reaction based methods (Fenton – Fe^{2+}/H_2O_2 and photo-Fenton (PF) – Fe^{2+}/H_2O_2/UV) [10,11].

Over the last two decades, electrochemical advanced oxidation processes (EAOPs) have gained increasing attention as a promising class of AOPs [12–15]. The former, simplest and most popular EAOP is anodic oxidation (AO), where organics can be directly oxidized at the anode surface by electron transfer and/or indirectly oxidized by $^{\bullet}OH$ weakly physisorbed at the anode surface and/or agents at the bulk solution such as active chlorine species, O_3, persulfates and H_2O_2 [16,17]. When AO is performed along with cathodic electrogeneration of H_2O_2, the process is called anodic oxidation with electrogenerated H_2O_2 (AO-H_2O_2) [18]. The electrochemical production of H_2O_2 with the addition of Fe^{2+} to the bulk originates the common and widely studied electro-Fenton (EF) process, in which additional $^{\bullet}OH$ are produced in the bulk from Fenton's reaction. Furthermore, Brillas' group has proposed and extensively studied the photoelectro-Fenton (PEF) and solar photoelectro-Fenton (SPEF) processes, which combine the EF technique with irradiation provided by artificial light or natural sunlight, respectively [13]. Other EAOPs like peroxi-coagulation (PC), Fered-Fenton, electrochemical peroxidation and sonoelectro-Fenton have also been applied to the remediation of various wastewaters [19–21]. Due to the high capital and operating costs of EAOPs, the development of combined treatment strategies including biological processes, chemical coagulation, electrocoagulation (EC) and membrane processes have also been proposed to optimize the wastewater treatment [19,22–24].

Recently, Sirés et al. [20] exposed a general overview on the application of AO, EF, PEF, SPEF and sonoelectrochemical processes for the treatment of synthetic and real wastewaters, focusing on the most updated works and giving a look to the future, but without an exhaustive and critical analysis of treatments. Other reviews have focused mainly on the use of technologies like AO, EF, sonoelectro-Fenton, Fered-Fenton and/or electrochemical peroxidation for the remediation of different kinds of effluents [5,14,17,19,25–28]. Vasudevan and Oturan [29] reviewed the use of distinct elec-