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Combination of advanced oxidation processes and active carbons adsorption for the treatment of simulated saline wastewater

Salvatore Cataldo [a], Adriano Iannì [b], Vittorio Loddo [b,*], Eliana Mirenda [a], Leonardo Palmisano [b], Francesco Parrino [b], Daniela Piazzese [c]

[a] Dipartimento di Fisica e Chimica (DiFC), Università degli Studi di Palermo, Viale delle Scienze ed. 17, I-90128 Palermo, Italy
[b] Dipartimento di Energia, Ingegneria dell'Informazione e Modelli Matematici (DEIM), Università degli Studi di Palermo, Viale delle Scienze ed. 6, I-90128 Palermo, Italy
[c] Dipartimento di Scienze della Terra e del Mare (DiSTeM), Università degli Studi di Palermo, CoNISMa-Palermo, Via Archirafi, 26, 90123 Palermo, Italy

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ABSTRACT

In the present work, an integrated process combining three different technologies, i.e. heterogeneous photocatalysis, homogeneous ozonation and adsorption on granular activated carbon (GAC) was used for the treatment of a synthetic solution simulating saline wastewater.

The synergistic effects between the different methods enhance the oxidation rate of the organic compounds in the integrated process, so that its use may result sustainable.

It has been found that a relatively low oxidation rate is sufficient to obtain the highest synergy between the three technologies and to maximize the performances. In particular, coupling ozonation and photocatalysis resulted in 20% enhancement in terms of reaction rate with respect to the sum of the rates of the single technologies. The contemporary presence of GAC and photocatalytic ozonation led to further enhancement of the whole system efficiency, obtaining a reaction rate 15% higher than the sum of the rates of the above mentioned single technologies.

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1. Introduction

One of the main source of marine degradation is represented by the disposal at sea of oily waste water, generated primarily by the washing operations of the ship's fuel tanks containing petroleum products (slops) and bilge water which originates from seawater infiltrations and leakage of engine oil and fuel. The discharge into the marine environment of oily wastewater is an issue of considerable technical and scientific interest, in relation to the toxic effects that such substances may have on the environment and human health. For these reasons, the International Maritime Organization (IMO) enacted the MARPOL 73/78 convention in which effluents from ships are limited to a maximum oil content of 15 ppm [1]. In order to preserve marine resources, the current legislation [2,3] has classified the oily wastewater generated from the naval transport, as special and hazardous waste, consequently imposing stringent rules for its disposal.

Water detoxification can provide a substantial contribution to water conservation, which is one of the primary objectives of sustainability. In fact, contaminated water can be recovered and utilized (or reutilized in a closed system) with vital savings of water resources.

Physical processes are now the most commonly used techniques to overcome this problem. Ships are equipped with oil-water separators which allow the separation of large amounts of oil. However, their efficiency results often inadequate because, in the presence of sediments, they may discharge at sea amounts of oil greater than 15 ppm. Moreover, the separation process is very time demanding so that the irreversible dissolution of organic compounds can be promoted and formation of emulsions may occur in the presence of detergents [4]. Among the physical methods, adsorption on activated carbons [5–7] has been proposed. Activated carbons are able to produce effluents of high quality, with amounts of oil lower than 10 mg L^{-1} and very low values of dissolved organic substances. Nevertheless, this physical treatment does not afford pollutant mineralization but only separation causing serious problems for their disposal [5]. Furthermore, carbons require to be substituted or regenerated once saturated. Recently, renewed attention has been paid to develop effective processes for removal of the contaminants present in water solution with high salt content (i.e. 35 g kg^{-1}) [8–10].

The use of advanced oxidation processes (AOPs) allows the mineralization of oils and pollutants present in this kind of

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* Corresponding author.
E-mail address: vittorio.loddo@unipa.it (V. Loddo).

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wastewater. Among the AOPs, heterogeneous photocatalysis has received in the last decade growing interest as it is an unselective oxidation process, which has been successfully used to oxidize many organic pollutants present in aqueous systems [11–15].

TiO_2 semiconductor is widely used as the catalyst for the photooxidation of organic compounds because it is inexpensive, (photo)-chemically stable and innocuous but also because it can be used both as polycrystalline powder and film onto suitable supports. On the other hand, photocatalysis alone suffers of very slow purification rates for high salinity solutions due to the presence of Cl^- ions and to the low O_2 concentration in solution because of the high ionic strength of the reacting medium.

Also ozonation has been successfully applied to accomplish this task [16,17]. Ozone (O_3) is one of the most powerful oxidants and it is used to aid the mineralization of organic contaminants from wastewater [17]. O_3 in combination with a semiconductor catalyst and near-UV radiation has been successfully used for the efficient treatment of organic contaminated waters [18–24].

However, ozonation of seawater presents a major drawback. Seawater contains Br^- ions (ca. 67 mg L^{-1}) which are almost quantitatively oxidized to BrO_3^- ions in the presence of O_3. BrO_3^- was classified as carcinogenic species from the IARC (International Agency for the Research on Cancer) and the USEPA (United States Environmental Protection Agency) and its minimum lethal concentration was fixed at 10 μg L^{-1}. TiO_2 photocatalysis was proposed [25] as a tool to overcome this problem. In fact, the electrons photogenerated under UV irradiation are able to reduce BrO_3^- to Br^- ions being, at the same time, the back reaction hindered.

For these reasons, we propose an integrated treatment system, obtained by combining ozonation and photocatalysis with an adsorption process on activated carbons, in order to ensure the simultaneous removal and degradation of hydrocarbon contaminants contained in polluted wastewater with high salinity. The complexity of bilge water has been simplified by using a synthetic sea water containing a recalcitrant probe molecule as 4-nitrophenol (NP) in the presence of heavy metal ions. The advantage of combining different technologies is the overcoming of the drawbacks of the methods used separately, the increase of the performance and, possibly, the reduction of the operating costs.

2. Experimental

The following sets of experimental runs were carried out: (i) adsorption onto granular activated carbons; (ii) heterogeneous photocatalysis by TiO_2 with the catalyst supported on glass beads; (iii) homogeneous ozonation; (iv) combined photocatalysis-ozonation process; (v) combined activated carbon-photocatalysis-ozonation process.

4-Nitrophenol (99.5%, Fluka), TiO_2 Aeroxide P25 (Evonik Industries), titanium tetraisopropoxide (97%, Sigma Aldrich), hydrofluoric acid (48% in H_2O, Sigma Aldrich), 2-propanol (Sigma Aldrich), triethanolamine (Riedel-de Haën), lead nitrate (Sigma Aldrich), zinc chloride (Carlo Erba), aluminum nitrate (Fluka), copper nitrate (Sigma Aldrich) and iron nitrate (99%, Sigma Aldrich) were used without further purification. Sodium chloride, supplied by Sigma Aldrich with a purity higher than 99%, was always dried at 110 °C before use. Filtrasorb 400 granular activated carbon (GAC) produced by Calgon Carbon Corporation (pore size ranging from 0.55 to 0.75 mm) was used without preliminary treatment for the removal of organic contaminants by water and wastewater. All solutions were prepared using milliQ pure water and class A glassware.

The experimental runs were carried out by using a solution simulating the naval wastewater. 4-Nitrophenol was chosen as a model pollutant since (i) the mechanism of its photocatalytic degradation is well known in literature, (ii) it belongs to the class of phenols which are present both in slops and bilge water, (iii) it is recalcitrant to advanced oxidation processes due to its aromaticity. Solutions of NP have been prepared by using both distilled and synthetic sea water. Distilled water solutions were used in order to determine the influence on the kinetics of (i) dissolved oxygen concentration, (ii) initial NP concentration, (iii) incident photon flux. Subsequently, other experimental runs were carried out by using solutions simulating real saline wastewater containing different initial NP concentrations. The amount of NP used in the synthetic wastewater ranged between 50 and 1158 mg L^{-1} and 33 g L^{-1} of NaCl were added in order to obtain the salinity of marine water. NP analyses were performed by a Shimadzu UV-2401 PC UV-vis spectrophotometer by measuring the intensity of the 315 nm absorption band. No interference of intermediate products was found. TOC analyses were performed with a Shimadzu analyzer (model TOC 5050A) coupled with an auto-sampler Shimadzu ASI-5000A to get information on the mineralization of the substrate. The artificial wastewater was prepared on the basis of the results obtained from the chemical characterization of different real samples or slops (S1-S5) collected from coastal storage of petroleum products (see Table 1). For the sake of brevity we report the analytical determinations only of 5 waste solutions, being similar all of the real samples analyzed. Results reveal moderate variability both for inorganic and organic contaminant concentrations, which mainly depend on the pre-treatment processes used (coagulation/flocculation or physical oil separation). The determined Cl^- ion content is similar for all of the samples and corresponds to the typical concentration in seawater (ca. 0.56 M).

Among the metal ions detected in the real samples, by using an ICP-OES Perkin-Elmer Optima 2100 coupled with an autosampler AS-90, an appropriate selection has been made by taking into account their abundance and toxicity, so that Pb^{2+}, Al^{3+}, Cu^{2+}, Zn^{2+}, Fe^{3+} have been chosen as the inorganic contaminants in the simulated wastewater at the concentration of 15 mg L^{-1} each. The precursors of the metal ions used were the corresponding nitrate salts. The pH values of the solutions ranged between 4 and 5 depending on the substrate concentration. Such pH values are not far from the corresponding values in real polluted effluents. Indeed, although fresh seawater presents pH values of ca. 8, due to the presence of carbonate buffers, pH values around 3 have been detected in polluted environments [26]. It is worth to note that the presence of metal ions is considered hereby as an input parameter which will be not discussed throughout the paper due to the

Table 1
Chemical characterization of five real sea wastewater samples taken from coastal storage of petroleum products. The figures are reported as mg L^{-1}.

Analytes	S1[a]	S2[b]	S3[a]	S4[b]	S5[a]
Pb(II)	0.009[c]	0.069	0.078	0.098	0.046
Al(III)	–	0.793	0.081	0.715	0.058
Cd(II)	0.004	–	–	–	0.002
Cu(II)	–	–	–	0.158	0.048
Cr(III)	0.007	0.008	0.008	0.013	0.021
Co(II)	0.005	0.007	0.006	0.005	0.005
Mn(II)	0.058	0.048	0.047	0.023	0.028
Ni(II)	0.011	0.008	0.014	0.111	0.113
Fe(III)	0.045	0.055	0.249	1.607	2.733
Zn(II)	0.025	–	0.137	2.121	1.973
Ga(II)	0.041	0.035	0.028	0.019	0.019
TOC[d]	74.70[e]	86.20	85.00	101.00	79.55

[a] Physical oil separation process as pre-treatment.
[b] Coagulation/flocculation process as pre-treatment.
[c] Standard error on metal ions measurements: ±0.002 mg L^{-1}.
[d] Total organic carbon.
[e] Standard error on TOC measurements: ±0.05 mg L^{-1}.

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