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Synergetic effect of gliding arc discharge treatment and biosorption for removal of nitrophene and glycine from aqueous solution

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ABSTRACT

The degradation of simulate house effluent containing Nitrophene and Glycine was carried out using gliding arc plasma reactor, one of the upcoming advance oxidation techniques. Aiming to reduce energy cost of treatment, the effect of various parameters such as treatment time, post-discharge phenomenon and addition of Jatropha natural biomass have been discussed. The experiments were conducted at a gas flow rate of 800 L/h. The degradation rate depends on the flow of highly oxidizing species formed in the plasma arc and on the sorbent/liquid contact surface time. Furthermore, the kinetic process of the in-situ treatment follows the pseudo-first order model while the post-discharge phenomenon is governed by the general order kinetic model. The initial high polluted solution with 950 mg/L TOC is degraded to 658 mg/L as final TOC after 1 h of in-situ treatment and to 460 mg/L TOC after 6 h in post-discharges. The use of Jatropha shell as biosorbent in addition of the plasma treatment reduces the TOC from 950 to 530 mg/L for in-situ treatment and, to 280 mg/L after 6 h in post-discharge. These results have showed that it is possible to significantly improve the efficiency of plasma treatment by incorporating modified agricultural residues. The occurrence of CO_2 is detected during the plasma treatment by Dräger tube analyser, while NO_2^- and NO_3^- are also identified by colorimetric tests.

1. Introduction

Water pollution is becoming an emerging challenge in developing countries. Many methods of water treatment exist, some as biological and physicochemical treatments require a lot of space and are expensive. This has led many scientists to propose alternatives techniques, for example transferring the pollutant from the aqueous phase to a solid phase e.g.: adsorption, coagulation and precipitation which have been longer satisfactory. But these techniques became ineffective in recent years, due to the exponential increase in the amount and complexity of discharged pollutants; the formed sludge treatment became more complex challenge. This is why recent works do not only focus on the removal of the contaminant from the aqueous solution but also on the techniques that will directly mineralize the pollutant molecule [1–3].

The present is based on the degradation of the nitrogen containing compounds such as 2,4-dinitrophenol (nitrophene) and glycine (Gly). These compounds are commonly found in effluents from food processing industries and have a direct impact on the quality of surface water and can lead to eutrophication.

The Gliding Arc (GA) discharge in humid air can offer an effective remediation solution in this case, due to its high oxidizing properties. The glidarc technique was firstly proposed for the treatments of gas, by Czernichowski and co-workers [4,5]. This technique has been extended to liquid treatments by several researchers [6–9]. Depending on the nature of the gas used, it is possible to produce directly new and very reactive chemical species and for discharges in moisten air, Benstaali et al. [10] showed that the major species were radicals HO• and NO•. This result revolutionized the plasma chemistry since the HO• radical in the presence of H_2O has a chemical potential of the order of E°

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Table 1
Composition of simulate wastewater.

Composition of main organic compounds			Auxiliary chemical	
Name and formula	Concentration (mg/L)	pKa	Name	Proportion **(v/v)**
Glycine: $H_2N.CH_2.COOH$	50	2.4 9.7	Mineral Acid (HCl 0.01 M and H_2SO_4 0.01 M)	5
			Ethanol	15
Nitrophene: $C_6H_4N_2O_5$	50	4.1	Distilled water	80
pH = 5.0; V = 450 mL				

(HO/H_2O) = 2.85 V/SHE, well above the potential of conventional oxidant such as $KMnO_4$, $K_2Cr_2O_7$, or even H_2O_2 and Cl_2. Plasma technology use electricity as the energy source and is environmental friendly because there is no subsequent sludge production. The major chemical properties expected for a gliding arc treatment of organic compounds are oxidizing and acidifying effects (when the target is not basic too much). Other properties of $ONOO^-$, i.e., nitrating, nitrosing effects and substituent effects of •OH should be considered as recommended by Nataili et al. [11]. The chemical properties of •OH, H_2O_2, NO and its derivatives ONOOH/$ONOO^-$ were carefully considered elsewhere [12]. We must only remind that nitric acid HNO_3 is a strong acid, and completely dissociated. Its oxidizing properties are not essential as compared to those of •OH [E°(OH/H_2O) = 2.85 V/SHE], H_2O_2 [E°(H_2O_2/H_2O) = 1.76 V/SHE] and above all ONOOH and its matching base peroxynitrite [E°(ONOOH/NO_2) = 2.05 V/SHE and E°($ONOO^-$/NO_2) = 2.44 V/SHE].

Besides most of the short plasma treatments are appreciated by countries which are short of electric energy because they can improve the natural properties of some agricultural residues, for example the removal power of biosorbent materials, which are otherwise only solid wastes. Recently, Prola et al. [13] have shown that the efficiency of biomass exposed to gliding arc plasma was significantly improved on removing a dye from aqueous solutions. The absorbing properties of *Jatropha curcas* shell (JN) were increased by exposure to non-thermal plasma. The JN biosorbent is a fibrous material with some macropores (pore with $\emptyset > 50$ nm). The plasma treatment changed the textural appearance of the yielding material, obtaining a biomaterial in form of thin lignocellulose sheets. Plasma treatment of *J. curcas* shell promoted an increase in the macropore structure on the biomaterial, contributing to a faster diffusion of the reactive red 120 dye (RR-120) through the pores of the sorbent.

The present work retains the same approach for confirmation purpose and the removal of Glycine.

($H_2NCH_2COOH \oplus H_3NCH_2COO^{\ominus}$) and Nitrophene (2,4-dinitrophenol) pollutants in simulate house wastewater was examined.

2. Experimental

2.1. Preparation of simulate waste water

A synthetic nitrogen containing house effluent involving glycine (amino-acid model) and 2,4-dinitrophenol (Nitrophene) is a suitable representative of usually found effluents of oil production and their corresponding auxiliary chemicals industry. A stock solution was prepared at pH 5.0. According to the practical information obtained from oil refinery effluents, the concentrations of organic compound and auxiliary chemicals are given in Table 1.

Glycine (NH_2–CH_2–COOH) was provided by BDH Biochemical Ltd., Poole England (99%). It is the simplest amino acids (the smallest), and the only one not to have rotatory power. Nitrophene is a toxic and flammable chemical; it has a high refractive index, and is slightly soluble in water, but has a good solubility in alcohols, ether and benzene. Nitrophene was provided by May&Baker Ltd Dagenham England (>99.0%).

2.2. Preparation of biosorbent

The *J. curcas* shell used in this work was provided by the Institute of Agricultural Research for Development (IRAD), Garoua-Cameroon. Following the published protocol of Prola et al. [13], the *J. curcas* shells (JN) used were washed with tap water to remove dust and with de-ionized water. These samples were then dried at 70 °C in an air-supplied oven for 8 h before being ground in a disk-mill and subsequently sieved (diameter particles ≤106 μm) [14]. The J.N biomaterial was now ready for plasma treatment according to the following procedure: 50.0 g of natural Jatropha was suspended in 500.0 mL deionized water disposed normally to the axis of the water cooled glass reactor (Fig. 1) at a distance of about 50 mm from the electrodes tips. The suspension was magnetically stirred and exposed to the plasma for t^* = 30 min (Gas flow: 800 L/h). The plume of the quenched plasma licks the liquid target and its components can then react at the surface or dissolve in the solution. After the discharge was switched off, the exposed mixture was centrifuged at 3600 rpm for 10 min, and the resulting biomass (JP) was washed several times with deionized water and dried at 70 °C in the oven until constant weight. After, JP was kept in glass bottle for further use.

2.3. The gliding arc plasma reactor

Fig. 1 shows the overall scheme of the experimental setup. The Gliding Arc (GlidArc) devices are a relatively new way (1988's patent) of cold plasma generation. The gliding arc apparatus is obtained by applying an alternate or continuous potential difference (pd) between two or several electrodes of continuously diverging profile [4]. The electrodes are laid out symmetrically around a gas jet (humid air). An electric arc occurs at the minimum spacing between electrodes and slips along the latter, thus generating excited species and free radicals which confer particular chemical properties to the plasma. This plume of soaked plasma licks a target and its species react at the target-plasma

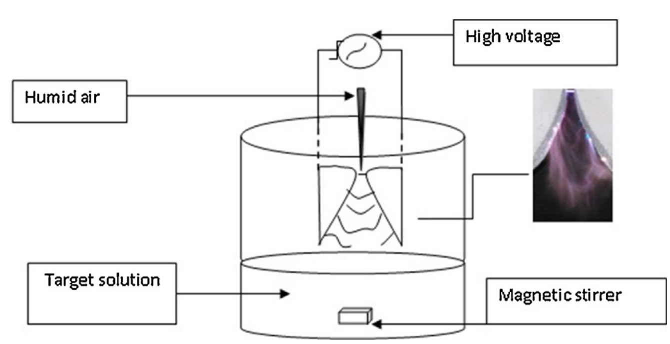

Fig. 1. Scheme of the experimental set-up involving a High Voltage transformer (HV = 10 kV/1A in open; estim. 600 V/160 mA in working conditions); electrode tips to target distance d = 2.5 cm. Gas flow rate: 800 L h^{-1}.