

Technical Note

Organic pollutants abatement and biodecontamination of brewery effluents by a non-thermal quenched plasma at atmospheric pressure

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Abstract

A preliminary study was carried out to investigate possible decontamination of the waste waters from a Cameroonian brewery by using a non-thermal atmospheric plasma. Samples of waste waters collected during periods of intense activity of production were first analysed for organoleptic, physical–chemical and organic parameters, and then exposed to a humid air electric discharge at ambient temperature and pressure. The resultant pollution abatement and germicidal effects are both ascribed to strong oxidising effects of OH and NO radicals formed in the plasma. The BOD removal efficiency of the process with brewery industrial wastewaters under BOD concentration of 385 and 1018 mg l⁻¹ were 74% and 98% respectively. The alkaline wastewaters are rapidly neutralised due to the pH lowering effects of the plasma treatment.

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1. Introduction

Plasma treatments are more and more often involved in the industrial processes of materials for many economical reasons: they allow much faster reactions to take place and need no use of any solvent nor catalyst. However, the use of electric discharges as suitable sources of plasma gases now enlarges to the treatment of gases and liquids in the scope of decontamination processes. The interactions between the constituting activated species in plasma and liquid phases are of fundamental interest to anticipate the results of a given treatment. The species created in an electric discharge in humid air, such as OH and NO radicals, present strong oxidising character so that they are able to participate in pollution abatement processes. Laboratory

experiments based on using gliding discharges for degrading spent solvents or organic solutes dispersed in water are numerous and concern various types of model molecules, contrary to the plasma treatments of actual industrial effluents. A pertinent example is the treatment of dye containing effluents from a textile plant (Abdelmalek et al., 2004). The electric discharges can then be considered as a particular advanced oxidation process and are actually efficient in pollution abatement of liquid or gaseous effluents. Additionally, the lethal effect of the discharges on bacteria has been recognized and studied (Laroussi et al., 2003; Moreau et al., 2005). This paper reports on using a humid air plasma provided by an electric gliding discharge in humid air to lower the pollutant concentration in brewery wastewaters.

1.1. The gliding discharge

The electric discharge selected for this study is a gliding discharge. The original device was first suggested by

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Lesueur et al. (1988). Czernichowsky (1994) developed the device with the same co-workers for volatile organic compounds abatement in industrial gaseous effluents. The device was developed in Rouen (France) and later in other laboratories, e.g., Mostaganem (Algeria), Tallahassee (USA), Zhejiang (China) or Yaounde (Cameroon) in view of degrading organic compounds in liquid phase, i.e., both spent organic solvents (Moussa and Brisset, 1999) and harmful organic solutes (Moussa et al., 2006). A next promising step for the development of the technique lies in inactivation of bacteria (Moreau et al., 2005); the technique was found more efficient (Hnatiuc, 2002; Doubla et al., 2005) than other kinds of electric discharges, such as corona or dielectric barrier discharges.

The gliding discharge is considered by many scientists as a convenient source for the generation of plasma at atmospheric pressure. When two diverging conductors (i.e., the electrodes), placed in a suitable gas (e.g., humid air) at a few mm apart, are connected to a convenient electric source, an electric arc forms at the shortest electrode gap and presents the character of a thermal plasma. The constituting electrons and heavy particles (molecules, ions and atoms) have then close energies and a local thermodynamic equilibrium is established. A gas flow placed along the reactor axis, gently pushes the feet of the arc along the electrodes so that its length increases until the arc is short-circuited by a new one and breaks into a plume. This plume sweeps the liquid surface with no contact allowed between the electrodes and the liquid target. The energy of the heavy particles then lowers, as does the macroscopic temperature, and a quenched plasma at quasi-ambient temperature and atmospheric pressure results in the plasma plume. The thermal effect is limited: for example, the temperature of a 200 ml sample of distilled water disposed in a non-thermostatted vessel never gets higher than 350 K after 1 h treatment.

1.2. Chemical reactions involved in a humid air plasma

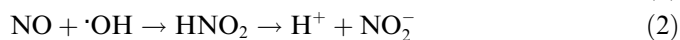
Exposure of aqueous solutions to such a plasma induces many reactions of the impinging activated species of the plasma with the solute molecules at the gas/liquid interface (Doubla et al., 2003; Abdelmalek et al., 2004).

The chemical reactions in solution are usually classified into acid–base, oxidation–reduction and complex formation reactions depending on the nature of the species exchanged (either protons, electrons or ligands respectively). The reactions involving radicals may be considered as a special case of electron exchange reactions and occur in substitution or addition processes: they have to be considered in the degradation mechanisms of organic targets.

The major reactions observed in an aqueous target exposed to a humid air plasma are lowering pH and strong oxidising effects. They are interpreted as a direct consequence of the formation of OH and NO radicals, the major reactive species identified and quantified in the plasma

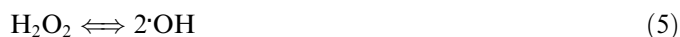
plume by spectrometry studies (Benstaali et al., 1998; Delair et al., 2001) and recently confirmed (Yan et al., 2005).

Short life species formed in the discharge may also react in the gas phase with the feeding gas molecules O₂, N₂, and H₂O mainly and yield a variety of oxygen and/or nitrogen containing species (e.g., ·O₂H, H₂O₂ and NO_x and their derivatives, but O₃ was not detected, probably due to the occurrence of water vapour which does not favour its formation). These reactive species are carried out to the liquid surface by the feeding gas where they can react with the solute molecules. Interaction between the electron flux of the discharge and water vapour may also be a source of reactive species such as the ·OH radicals in a way similar to that occurring in water radiolysis, and be responsible for the acid effects:



The increasing concentration of the solvated protons matches those of the solvated nitrite and nitrate ions. Arguments to support this simple model are provided by the analysis of nitrite and nitrate ions in plasma treated aqueous solutions (Marouf-Khelifa et al., 2006). The NO₂⁻ concentration presents a maximum for increasing plasma treatment time, while that of NO₃⁻ continuously increases. The evolution of NO₂⁻ to NO₃⁻ may be connected with the thermodynamic stability of N(II), N(III) and N(V) compounds: the Pourbaix diagrams show that NO₂⁻ is unstable in acid medium (pH < 2.6) and disproportionates to NO₃⁻ and NO. The nitrite ions are thus a good example of long life transient species formed in the plasma treatment of aqueous solutions. Incidentally, the influence of NO_x in acidifying the aqueous target is backed up by measurements performed after plasma treatments in humid oxygen which induce no significant pH lowering.

The ·OH radical is a very powerful oxidising agent [$E^0(\cdot\text{OH}/\text{H}_2\text{O}) = 2.85 \text{ V/NHE}$] and thus responsible for oxidation reactions with organic targets, both due to its own properties and to its derivative and/or parent molecule H₂O₂ (Eq. (5)).



NO leads first to the formation of nitrite in neutral medium, which is further oxidised to nitrate ions as stable species, thus associated with acid changes. The acid effect is detailed elsewhere (Benstaali et al., 1998; Moussa et al., 2005) and readily occurs. Additionally, the high standard oxidation–reduction potentials of the HNO₂/NO (1.00 V) and NO₃⁻/HNO₂ (1.04 V) systems reflect the oxidising power of the nitrate ion.

This innovative technique was successfully used for oxidising many compounds such as iron(II) complexes (Doubla et al., 2003), azoic dyes in textile effluents (Abdelmalek et al., 2004), spent solvents (Moussa and Brisset, 1999,

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