



Microwave atmospheric pressure plasma jets for wastewater treatment: Degradation of methylene blue as a model dye



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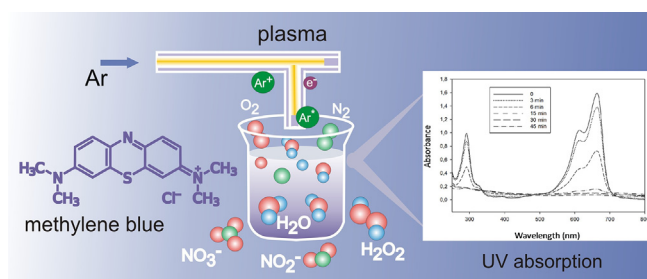
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HIGHLIGHTS

- Microwave plasma jets were applied for MB degradation in water.
- Degradation of MB is related to H₂O₂ formation.
- OES techniques were used to identify plasma active species triggering AOPs.
- This plasma-over-liquid configuration provided good reaction rates and energy yields.

GRAPHICAL ABSTRACT



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ABSTRACT

The degradation of methylene blue in aqueous solution as a model dye using a non thermal microwave (2.45 GHz) plasma jet at atmospheric pressure has been investigated. Argon has been used as feed gas and aqueous solutions with different concentrations of the dye were treated using the effluent from plasma jet in a remote exposure. The removal efficiency increased as the dye concentration decreased from 250 to 5 ppm. Methylene blue degrades after different treatment times, depending on the experimental plasma conditions. Thus, kinetic constants up to 0.177 min^{-1} were obtained. The higher the Ar flow, the faster the degradation rate. Optical emission spectroscopy (OES) was used to gather information about the species present in the gas phase, specifically excited argon atoms. Argon excited species and hydrogen peroxide play an important role in the degradation of the dye. In fact, the conversion of methylene blue was directly related to the density of argon excited species in the gas phase and the concentration of hydrogen peroxide in the aqueous liquid phase. Values of energy yield at 50% dye conversion of 0.296 g/kWh were achieved. Also, the use of two plasma applicators in parallel has been proven to improve energy efficiency.

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1. Introduction

In recent times there has been a dramatic increase in the amount of organic pollutants (volatile organic compounds,

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pharmaceuticals, organic dyes...) detected in water resources, thus representing a serious concern for the public health. Among these contaminants, dye wastewaters from the textile industry are particularly widespread (approx. 1 million kg of dyestuffs are emitted into the environment per year) (Foster et al., 2012) and pose numerous problems (affectation of photosynthesis in water plants, carcinogenicity, etc.). Consequently, the removal of colour from textile industry wastewaters represents a major environmental goal as it enables water reuse for further textile mill processing. However, dyes are designed to resist degradation and so this is not an easy task. Indeed, the new organic dyes coming to market have very stable molecular structures so conventional wastewater treatment techniques are usually ineffective for their degradation. As a consequence, new technologies for removal of dyes from wastewater have been investigated.

Advanced Oxidation Techniques and Processes (AOPs) are approaches allowing in situ decomposition of organic compounds in water through mineralization, i.e. conversion of the compound to carbon dioxide, water and inorganic intermediates (Glaze et al., 1987). AOPs have been developed to generate hydroxyl free radicals by different techniques (Al-Kdasi et al., 2004), and use these radicals as strong oxidants. More generally, the term AOP also refers to chemical processes and precursors that have high reduction potentials and either produce OH or directly attack organic molecules (ozone, atomic oxygen, excited nitrogen, ultrasound, UV light, and peroxide) (Foster et al., 2012). AOPs have been proven to be powerful and efficient treatment methods for degrading recalcitrant materials and toxic contaminants (Mohajerani et al., 2009).

In the last years, plasmas have begun to be employed to induce AOPs for water treatment purposes (Anpilov et al., 2001; Lukes et al., 2005; Sato and Yasuoka, 2008; Stratton et al., 2015). More specifically, plasmas are sources of excited species, charged particles, radicals and UV radiation, among others, each of which are themselves advanced oxidation techniques (Foster et al., 2012). Different kinds of plasmas, using different configurations have been employed for water (and other liquids) treatment (Malik, 2010), particularly for the abatement of organic pollutants such as dyes (Clements et al., 1987; Yang et al., 2005; Magureanu et al., 2007; Dojčinović et al., 2011; Reddy et al., 2013), phenolic compounds (Sun et al., 2000; Krugly et al., 2015) and antibiotics (Magureanu et al., 2011; Kim et al., 2015).

Nowadays non-thermal plasmas, i.e. those whose electron temperature is different from the corresponding to ions and neutrals (gas temperature), are considered as a very promising technology. Their non-equilibrium properties including low power consumption and their ability to achieve enhanced gas phase chemistry at relatively low gas temperature, are responsible for the great attention that these plasmas have gained from an applied point of view and their extensive use in applications that require low temperatures, including material processing and synthesis, biomedical applications, and surface modification, among others (Park et al., 2001; Selwyn et al., 2001; Zille et al., 2015).

In particular, non-thermal atmospheric-pressure plasma jets/plumes play an increasingly important role in various plasma-assisted applications for several reasons (Walsh et al., 2006; Laroussi and Akan, 2007; Laroussi, 2009; Walsh et al., 2010; Lu et al., 2012). First of all, due to their practical capability in providing plasmas which are not spatially confined by electrodes and whose stability is not compromised by the presence of the sample to be processed. Plasma jets are able to generate stable discharges in a gas and then flush it to a separate region of reactive gas for processing applications (Walsh et al., 2006). This spatial separation enables a considerable flexibility in jet designs in order to achieve a good control in both plasma dynamics and reaction chemistry (Mora et al., 2010, 2011). Secondly, because plasma jet

configuration offers a chamberless delivery of downstream reaction chemistry if desired (Walsh et al., 2010). Finally, they are free from constraints imposed by vacuum-compatibility, as they operate at atmospheric pressure, being this an undeniable advantage in practical applications.

Atmospheric plasma jets can be generated in many different gases and using different electrical excitations. Several cold plasma jet devices have been designed; most of them are described in Laroussi and Akan review (Laroussi and Akan, 2007) although there are others more recently developed such as the plasmas generated by using nanosecond dc voltage pulses with kilohertz repetition frequencies as well as with sine-wave excitations in the kilohertz-to-megahertz range in pulsed or continuous mode (Chandana et al., 2015).

In the present work, we report for the first time on the design of a plasma jet reactor allowing liquid (water) treatment based on the use of a microwave (2.45 GHz) surface wave sustained discharge at atmospheric pressure (Ferreira and Moisan, 1993). This type of discharge generates non thermal plasmas with a high electron density (in the order of 10^{14} cm⁻³), and so a high reactivity (Henriques et al., 2011b). Microwave discharge reactors enable the generation of very stable and reproducible plasmas, that can operate over a broad range of experimental conditions (pressure, gas type, frequency, power, geometry), so being capable to produce copious and controlled amounts of active species. As far as the authors know, this is the first study where a microwave plasma jet of this type have been employed for water treatment.

More specifically, the reactor consisted of a plasma-jet over liquid configuration that was employed for degradation of methylene blue in aqueous solution. Remote exposure of the sample was done (Laroussi, 2009). In contrast to direct exposure (in which the sample is subject to all possible agents generated by the plasmas including heat, charged particles, reactive neutrals, and electromagnetic radiation), in remote exposure the effect of charged particles on the water sample under treatment is weak as the sample is placed at some distance from the plasma and most of these species recombine before reaching it. The heat flux is also greatly reduced in this case, what leaves mainly the long-lived radicals to directly interact with the sample (Laroussi, 2009).

2. Experimental part

2.1. Chemicals

Methylene blue (MB) was selected as the model organic contaminant dye because of its stable molecular structure. Moreover, MB-laden waste water exhibit a high chroma and toxicity and has become an unmanageable industrial wastewater (Wang et al., 2013) whose treatment has being profusely studied in the last years. Methylene blue (Fig. S1) is a heterocyclic aromatic compound belonging to the phenothiazine family (C₁₆H₁₈N₃SCl). It was purchased from Sigma-Aldrich and used without further purification.

2.2. Plasma reactor

Fig. 1 shows the experimental set-up used for the generation of the plasma. A surfatron device (Moisan and Pelletier, 1992) was used to couple the energy coming from a microwave (2.45 GHz) generator (with a maximum stationary power of 200 W in continuous-wave mode) to the support gas (argon with a purity $\geq 99.995\%$) within a quartz reactor tube of 1.5 and 4 mm of inner and outer diameter, respectively, opened to the air as shown in Fig. 1. Surfatron was originally designed to generate cylindrical plasma columns inside straight dielectric tubes (Moisan and Pelletier, 1992), sustained by an azimuthally symmetric TM₀₀

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